THE FERMI SURFACE OF COPPER BY POSITRON ANNIHILATION

by

PETER PETIJEVICH

B.A.Sc., University of British Columbia, 1963

A THESIS SUBMITTED IN PARTIAL FULFILMENT OF

t

THE REQUIREMENTS FOR THE DEGREE OF

MASTER OF SCIENCE

in the Department

of

PHYSICS

We accept this thesis as conforming to the required standard

THE UNIVERSITY OF BRITISH COLUMBIA

September, 1966

In presenting this thesis in partial fulfilment of the requirements for an advanced degree at the University of British Columbia, I agree that the Library shall make it freely available for reference and study. I further agree that permission for extensive copying of this thesis for scholarly purposes may be granted by the Head of my Department or by his representatives. It is understood that copying or publication of this thesis for financial gain shall not be allowed without my written permission.

Department of PHYSICS

The University of British Columbia, Vancouver 8, Canada.

Date <u>Sept. 1966</u>

ABSTRACT

A study of the Fermi surface of copper at room temperature has been made by means of a positron annihilation technique.

A positron active copper single crystel was placed midway between two "point" scintiliation counters operated in time coincidence. The coincidence count rate was measured for various crystal orientations and the count rate interpreted as a measure of the diameter of the Fermi surface.

The experiment yields a Fermi surface that is spherical in \vec{k} -space except for protrusions in the {iii} directions which are estimated to subtend an angle of about 20° at $\vec{k} = 0$. Within experimental error the results are consistent with those obtained by other methods near 0° K.

ACKNOWLEDGEMENTS

The author wishes to express his gratitude to Dr. G. Jones for his helpful advice and kind supervision throughout the duration of this work.

Thanks are also due to Dr. D. Ll. Williams for his many helpful discussions and suggestions.

The author wishes to thank Dr. E. Teghtsoonian and Mr. N. R. Risebrough of the Department of Metallurgy for their assistance with the x-ray diffraction study of the crystal used in the present work.

Thanks are also extended to Professor S. H. de Jong of the Department of Civil Engineering for making available the engineer's transit used in this work.

The author also wishes to thank Mr. A. Fraser for his assistance with the design of the crystal rotation assembly.

TABLE OF CONTENTS

.

.

		Page
Chapter I	INTRODUCTION	1
Chapter II	MOTION OF ELECTRONS IN METALS	7
Α.	Introduction	7
Β.	The One-Electron Approximation	
С.	The Free-Electron Model	
D,	The Crystal Lattice	
E,	The Reciprocal Lattice	
F.	Motion of an Electron in a Crystal Lattice	18
	1. Perturbation Theory for Weak Periodic Potentials	20
	2. Effect of Electron Correlations on the Fermi Surface	22
	3. Justification of the One-Electron Approximation	22
	4. Energy Band Calculations by use of the One-Electron Model	n 23
	5. The Fermi Surface of Copper: Theory and Experiment	: 24
Chapter III	ANNIHILATION OF POSITRONS	
Α.	Introduction	
Β.	Free Annihilation of Positrons	
С.	Positron Annihilation from a Bound State	29
	Bound Electron-Positron Systems	29
	2. Positronium	30
	3. Positronium Annihilation	30
D.	Angular Correlation of Two-Photon Annihilation of Positrons	31

		Page
Ε.	Angular Correlation Geometries	33
	1. Introduction	33
	2(a). Wide Slit Geometry	33
	2(b). Determination of Fermi Surfaces by Wide Slit Geometry	35
	3(a). Point Geometry	37
	3(b). Determination of Fermi Surfaces by use of Point Geometry	38
	4. Collinear Point Geometry	40
F.	Annihilation of Positrons in Electron Gases	40
	1. Effect of Electron-Electron Interactions	40
	2. Effect of Electron-Positron Interactions	41
	 Effect of Electron-Electron and Electron-Positron Interactions 	41
	4. Comparison with Experiment	42
G,	Annihilation of Positrons in Real Metals	44
	1. Introduction	44
	2. Effect of the Periodic Crystal Lattice Potential	44
	3. Effect of Core Annihilation	45
Н.	Lifetimes of Positrons in Metals	45
Chapter IV	EXPERIMENTAL ARRANGEMENT	47
Α.	Metal Crystal and Positron Source	49
Β.	Orientation of Crystal	50
C.	Spatial Stability of Notch and Pin Assembly	52
D.	Crystal Holder	52
E.	Holder Support	53
F,	Gamma Counters	55
G.	Electronics	57
Н.	Stability of Electronics	57

.

		Page
Chapter V	RESULTS AND CONCLUSIONS	61
Α.	Introduction	61
В.	Experiment	62
C.	Results	
D.	Interpretation of the Results	65
	1. Effects of Finite Source and Detector Size	65
	2. Resolution Function for Finite Detectors and Point Crystal	66
	 Resolution Function for Finite Detectors and Finite Crystal 	67
E.	Interpretation of the Results	69
F.	Accuracy Attainable with the Method	70
	1. Resolution and Counting Rate	70
	2. Stability	71
G.	Discussion	72
Н.	Conclusions	73
APPENDICES		
Α.	SOLUTION OF ABEL'S INTEGRAL EQUATION	75
Β.	EXPECTED ANGULAR CORRELATION CURVE WIDTH	77
С.	EFFECT OF CORE ANNIHILATION	80
Bibliography		81

vii

LIST OF TABLES

Table	· ·	Page
I	Comparison of Theoretical and Experimental Fermi Surface	
	Dimensions	26

.

viii

LIST OF FIGURES

Figure		Page
۱.	Body-centered Cubic Structure	14
2.	Face-centered Cubic Structure	15
3.	Hexagonal Close-packed Structure	16
4.	Brillouin Zone for F.C.C. Lattice	19
5.	Copper Fermi Surface Details	25
6.	Two-Photon Annihilation	32
7.	Wide-slit Geometry	32
8.	Wide-slit Geometry Results	35
9.	Region Sampled by Wide-slit Geometry	35
10.	Point Geometry	38
11.	Region Sampled by Point Geometry	38
12.	Collinear Point Geometry	38
13.	Annihilation in Electron Gases	43
14.	Positron Annihilation Rates	43
15.	Experimental Arrangement	48
16.	Notch and Pin Arrangement	51
17.	Crystal Holder	54
18.	Remote Control	54
19.	Preamplifier and Shaper Circuit	58
20.	Coincidence Circuit	59
21.	Experimental Results	64
22.	Fermi Surface Neck Details	65

CHAPTER |

INTRODUCTION

One of the fundamental problems in the theory of the solid state is to find a solution of the Schrodinger equation for the many-body system defined by a crystalline solid. Since a direct solution of this problem is not possible considerable effort has been directed toward the production of models which approximate the many-body system. By the use of these models it is then possible to make predictions about the properties of the crystalline system that can be compared with those derived from experiment. It is thus desirable to be able to make reliable measurements of these properties so that they can be compared with the predictions of the various theoretical models and also because the data may be used to suggest newer and better models.

A special case of this problem that is particularly interesting occurs when the crystalline solid is a metal. Work on this aspect of the problem can be considered to have started with the free-electron approximation of Sommerfeld (1928) in which it was assumed that the electrons of the solid do not interact with each other or with the ion cores of the crystal lattice. Despite its oversimplification of the actual physical situation, the theory usually gives good qualitative results. However, in order that satisfactory quantitative results be obtained, it is usually necessary to include the effect of the ion cores. The extensive theoretical work on this aspect of the problem is essentially an extension of the studies of Hartree (1928), Bloch (1928), and Fock (1930). Inclusion of the effect of the ion cores greatly complicates the problem so that accurate calculations are difficult to perform, but it usually leads to results that are in better accord with the large body of available experimental data than does the simple Sommerfeld model. However, it is reasonable to assume that agreement with experiment would be further improved if it were possible to include the effect of electron-electron interactions. Considerable progress on this aspect of the problem has resulted from the early studies of Bohm and Pines (Pines, 1955; Pines, 1963). Most calculations of energy bands in metals at present are still based on the independent particle model (Chapter II), however, because of the difficulty of adequately including the effects of electron-electron interaction (Reitz, 1955; Callaway, 1958).

Since the work of Wigner and Seitz (1933) on sodium, well over a hundred band calculations for metals have been performed. In recent years the advent of high-speed computing machines has made possible an increasing number of extensive calculations that agree well with one another and with experiment. However, with a few exceptions, good accord with experiment is limited to metals of the alkali group or the alkaline earth group (Callaway, 1958).

An exception that is of particular interest is copper. Here the agreement of recent extensive band calculations with experiment is good (Segall, 1962: Burdick, 1963). The experimental situation is also rather unique since the electronic properties of copper have been more thoroughly investigated than those of any other metal (Segall, 1962). For example, the Fermi surface which describes the highest occupied electron energy level at absolute zero, has been studied by at least five major experimental techniques (Harrison and Webb, 1960). These include the methods of anomalous skin effect, magnetoresistance, magnetoacoustic effect, cyclotron resonance, and the de Haas-van Alphen effect. All five methods give results that are in accord with the model of the Fermi surface for copper proposed by Pippard (1957). In this model the Fermi surface consists of a spherical central part (the "belly") together with eight protrusions ("necks") along the {111} directions. As discussed in Chapter II, this model for the Fermi surface of copper is in good accord with the surface predicted by recent band calculations (Segall, 1962; Burdick, 1963).

The experimental techniques mentioned above are all limited to use on fairly pure metal specimens at very low temperatures ($\sim 4^{\circ}$ K) due to the requirement of long electronic mean free path (Harrison and Webb, 1960). This requirement is rather restrictive. For example, it makes impossible a satisfactory study of alloys. The requirement of low temperatures complicates the study of metals such as sodium and potassium in which low temperature phase transitions occur. The restriction to low temperatures also eliminates the possibility of examining adequately the temperature dependence of the "sharpness" of the Fermi surface boundary. Finally, it is to be noted that in these methods one generally examines only the electron states that reside near the Fermi surface. It would be of considerable interest to be able to probe the entire valence band.

A technique that does not suffer from the above limitations involves a study of the annihilation of positrons in metals. In this method the metal to be studied is bombarded with positrons. Upon collision with an electron, there is a chance that the pair may annihilate each other. In nearly every such annihilation two nearly antiparallel photons result. The small deviation from strict antiparallelism ($\sim 10^{-3}$ radians) is related to the centre-of-mass momentum of the annihilating electron-positron pair by the simple relation $p_{+} = mc\Theta$ where p_{+} is the component of pair momentum perpendicular to the nearly antiparallel photon pair, m the electronic rest mass, c the speed of light, and \mathbf{e} the angular deviation from 180°. The energy of the photons emanating from the sample is about 0.51 Mev (annihilation radiation) so that gamma-ray attentuation and scattering by the sample will be negligible if the sample is reasonably small. Since calculations by Lee-Whiting (1955) indicate that the positron is essentially at rest (thermalized) before it annihilates, the centre-of-mass momentum of the pair is essentially that of the electron.

The angular correlation of the gamma-ray pairs emitted in positron annihilation in solids has been observed by many workers. The early work of Klemperer (1934) and that of Montgomery and Beringer (1942) established the time coincidence and near collinearity of the two gamma-rays emitted in positron annihilation. The first detailed angular distribution was obtained by De Benedetti et al. (1950). Since then angular correlation studies of many elements and compounds have been made. The subject has been reviewed by Wallace (1960).

Most of the work on Fermi surfaces by the positron annihilation

technique has used the "wide-slit" method (Chapter III). On the basis of the free-electron model it is easy to show that this wide-slit method samples a slice through the Fermi sphere (Chapter III). By the use of this technique it has been possible to obtain the momentum distribution of the electrons within a metal at temperatures ranging from $\sim 4^{\circ}$ K (Stump, 1955) to beyond the melting point (Gustafson et al., 1963), providing a rather direct verification of Fermi-Dirac statistics. However, due to finite resolution and other complicating factors (core annihilation and other higher momentum effects arising from the presence of the crystal lattice) which are discussed in Chapter III, the method has not been useful for yielding quantitative information about the detailed shapes of Fermi surfaces.

A "point geometry" method which samples a cylindrical volume in momentum space has been used by a few workers (Colombino et al., 1963; Fujiwara, 1965). Although this method permits improved resolution compared to the wide slit method, the results are again obscured by core annihilation and other lattice effects making it difficult to use the method for an accurate study of Fermi surface topology.

The work of this thesis describes a development of the "point geometry" technique which offers advantages for the study of Fermi surfaces. With this method (collinear point geometry") it is possible to examine the Fermi surface more quantitatively than appears possible by the "wide-slit" or "point geometry" methods because core annihilation and higher momentum effects arising from the presence of the crystal lattice play a relatively less important role in the new technique.

The principles of this new "collinear point geometry" technique are discussed in Chapter III of the present work. This chapter is followed

by Chapter IV in which the experimental arrangement is discussed in some detail. Finally, in Chapter V some experimental results obtained from an application of this new method to a study of the Fermi surface of copper at room temperature are presented. These results are used to construct a Fermi surface for copper which is then compared with the copper Fermi surface obtained at very low temperatures by the more precise conventional techniques mentioned above. The discussion closes with a statement of the conclusions that have been drawn from the present study.

CHAPTER II

MOTION OF ELECTRONS IN METALS

A. Introduction

If one considers a crystalline solid at fairly low temperatures so that the motion of the relatively massive nuclei can be neglected, one can to a good approximation (Ziman, 1960) write the Schrödinger equation for the system as

$$\begin{bmatrix} -\sum_{i=1}^{N^{2}} \frac{\pi^{2} \nabla_{i}^{2}}{2m} - \sum_{i=1}^{N^{2}} \frac{e^{2} z}{j = 1} + \frac{1}{2} \sum_{i=1}^{N^{2}} \frac{e^{2} z}{j = 1} \end{bmatrix} \Psi = E \Psi$$
(2-1)

Here it has been assumed that the lattice is perfect and that the crystal is composed of N atoms each with z electrons. The position vector of electron \dot{k} is denoted by \vec{r}_i and that of nucleus \vec{j} by \vec{R}_j . The remaining symbols have the usual meaning: -e is the electronic charge, m the electronic rest mass, $2\pi\hbar$ Planck's constant and E the energy eigenvalue for the system. The interpretation of the terms in the above Hamiltonian is well-known (Ziman, 1960). The first term represents the kinetic energy of the electrons, the second the potential energy of the electrons in the nuclear Coulomb field, and the third term the potential energy of the electron-electron Coulomb interaction. If more than one type of atom is present the Hamiltonian of equation (2-1) is easily generalized and leads to an expression that is only slightly more complicated.

B. The One-Electron Approximation

The above equation is quite general and is in fact one of the fundamental equations in the theory of solids. However, since in a typical solid one has a very large number of interacting particles it is quite impossible to solve the Schrodinger equation directly. Nevertheless the problem can be made more or less tractable if one treats the electrons as statistically independent (the one-electron approximation). The wave function for the system then becomes (Anderson, 1963)

$$\Psi(\vec{x}_1 \cdots \vec{x}_{N_z}) = \Psi_1(\vec{x}_1) \Psi_2(\vec{x}_2) \cdots \Psi_{N_z}(\vec{x}_{N_z})$$
(2-2)

where the $\gamma_{\ell}(\vec{x})$ are one-electron functions (including spin). If, in addition to the one-electron approximation, the electron-electron interaction term is replaced by its average value one obtains the Hartree equation with one-electron eigenvalue :

$$\begin{bmatrix} -\frac{\hbar}{2m} \nabla_i^2 - \sum_{j=1}^{N} \frac{e^2 z}{|\vec{r}_j - \vec{r}_i|} + \sum_{i \neq j}^{N^2} \int \frac{e^2 |\Psi_j(\vec{r}_j)|^2 d\vec{r}_i}{|\vec{r}_i - \vec{r}_j|} \psi_i(\vec{r}_i) = \epsilon_i \Psi_i(\vec{r}_i)$$
(2-3)

where i = 1, 2, ... Nz. In these equations the average interaction term has a simple interpretation if it is noted that since the charge density of electron j at point \vec{r}_{i} is -e $|\Psi_{i}(\vec{r}_{i})|^{2}$, the potential energy associated with electron i and volume element $d\vec{r}_{i}$ is $\frac{e^{i}|\Psi_{i}(\vec{r}_{i})|^{2}d\vec{r}_{i}}{|\vec{r}_{i} - \vec{r}_{i}|}$

Then the quantity

$$V_{i}(\vec{r}_{i}) = \int \frac{e^{2} |\psi_{i}(\vec{r}_{j})|^{2} d\vec{r}_{j}}{|\vec{r}_{i} - \vec{r}_{j}|}$$

can be interpreted as the potential energy of electron i in the charge cloud of electron j (Raimes, 1961).

In order to solve the Hartree equations one assumes a set of γ_i , calculates V_i (\vec{r}_i) and thence a new set of γ_i which are in turn used in the Hartree equations and so on, until a self-consistent result is obtained.

Since electrons are fermions the function \mathcal{Y} must be antisymmetric in the electron coordinates. However, the product function given by equation (2-2) does not obey this condition. However, it is possible to satisfy the antisymmetry requirement by forming a suitable linear combination of product functions; the Slater determinant, given by:

$$\Psi = \begin{bmatrix} (NZ)^{l} \end{bmatrix}^{-\frac{1}{2}} \qquad \begin{array}{c} \Psi_{i}(\vec{x}_{i}) & \Psi_{i}(\vec{x}_{2}) & \cdots & \Psi_{i}(\vec{x}_{NZ}) \\ \Psi_{2}(\vec{x}_{i}) & & \\ \Psi_{NZ}(\vec{x}_{1}) & \cdots & \ddots & \Psi_{NZ}(\vec{x}_{NZ}) \end{array}$$

which is antisymmetric as required. If for two electrons one has $\vec{x}_1 = \vec{x}_2$ (spatial and spin coordinates identical) the determinant vanishes, in accord with the Pauli exclusion principle.

Using the determinantal function it is possible to construct a better set of one-electron equations (Anderson, 1963). These are the Hartree-Fock equations:

$$-\frac{\pi}{2m}\nabla_{i\sigma}^{2}\psi(\vec{r}_{i}) - \frac{2}{j}\frac{e^{2}z}{|\vec{R}_{j}-\vec{F}_{i}|}\psi(\vec{r}_{i}) + \left(\sum_{j,\sigma'}\int\frac{e^{2}|\vec{\gamma}_{j\sigma'}(\vec{r}_{2})|^{2}d\vec{z}}{|\vec{F}_{i}-\vec{F}_{2}|}\psi(\vec{r}_{i}) - \left(\sum_{j}\int\frac{e^{2}\psi_{i\sigma}(\vec{r}_{2})\psi_{i\sigma}(\vec{r}_{2})d\vec{r}_{i}}{|\vec{F}_{i}-\vec{F}_{2}|}\right)\psi_{j\sigma}(\vec{r}_{i}) = \epsilon_{i}\psi_{i\sigma}(\vec{r}_{i})$$

where **c** denotes the spin label. Although these equations are known as the Hartree-Fock equations, they are actually a special case of a more general method (Messiah, 1962) that is of considerable importance in atomic (Hartree, 1957), nuclear (Brown, 1964), and solid state problems (Anderson, 1963). However, in what follows, the discussion of the method will be limited to its use in the theory of metals.

Due to their mutual Coulomb repulsion, the electrons in a solid will tend to avoid each other (Coulomb correlation). Thus the electrons will not move independently of each other as is assumed in the one-electron model. In the Hartree and Hartree-Fock methods the Coulomb correlation of the electrons is not taken into account. In the Hartree-Fock method, however, the Pauli exclusion principle does introduce a correlation between electrons of like spin. In general, inclusion of the Coulomb correlations and spin correlations will reduce the energy of a many-electron system because the correlations are effective in making the electrons spend less of their time near each other. In a treatment of metals by the one-electron method it is often found better to ignore all correlations than to include only some of them (Raimes, 1961). Thus the Hartree-Fock method.

Despite its limitations, the Hartree method has often been used in the theory of metals (Reitz, 1955). In particular, it can be used to derive the properties of a free-electron gas. This is particularly useful since a free-electron gas can be regarded as a crude model of a metal.

C. The Free-Electron Model

If one replaces the charge distribution of the ion cores by a

uniform distribution of positive charge, and the charge distribution of the valence electrons by a uniform distribution of negative charge so that the net charge is zero, the Hartree equation simplifies to

$$-\frac{\hbar^2}{2m}\nabla^2\psi = \mathcal{E}\Psi$$
(2-4)

For a cube of side L, containing N electrons it is seen that a solution of (2-4) is

$$\Psi(\vec{r}) = \frac{1}{\sqrt{L^2}} e^{i\vec{K}\cdot\vec{r}}$$
(2-5)

provided that

$$\epsilon(\vec{k}) = \frac{\vec{k} \cdot \vec{k}^2}{2m}$$
(2-6)

Application of the usual periodic boundary conditions (Ziman, 1960; Raimes, 1961): $\Psi(x+L,y,z) = \Psi(x,y+L,z) = \Psi(x,y,z+L)$ then yields

$$\vec{k} = \frac{2\pi}{L} (n_1 \hat{i} + n_2 \hat{j} + n_3 \hat{k})$$
 (2-7)

where the n_i are positive or negative integers or zero and \hat{i} , \hat{j} , \hat{k} are unit vectors along the cube edges.

From (2-7) it is seen that the integers n. representing "orbital" states (Raimes, 1961) define a lattice in \vec{k} -space. Since each cube of side $\frac{2\pi}{L}$ will contain one such orbital state, the number of orbital states per unit volume of k-space is $\frac{L^3}{8\pi^3}$. Thus in a volume element $d\vec{k}$ of \vec{k} -space there are $\frac{L^3}{8\pi^3}d\vec{k}$ orbital states. Generalizing slightly, it is seen that for a metal of volume v there are $\frac{2vdk}{8\pi^3}$ electron states (spin degeneracy included) in a volume element $d\vec{k}$ of \vec{k} -space.

It is evident from (2-6) that the surfaces of constant energy are spheres. Thus, as a consequence of Fermi-Dirac statistics, the occupied

region of k-space at the absolute zero of temperature will be a uniformly dense sphere. Denoting the radius of this sphere (the Fermi sphere) by k_{p} and observing that there are N electron states within the Fermi sphere the following condition is obtained:

$$\frac{(2\mathbf{v}) \frac{4}{3}\pi k_{F}^{3}=N}{k_{F}^{2}=\left[\frac{(3\pi N)}{(\mathbf{v})}\right]^{\frac{1}{3}}}$$

and the Fermi energy is

$$\epsilon_{F} = \frac{\hbar^{2} k_{F}^{2}}{2 m} = \frac{\hbar^{2}}{2 m} \left(\frac{3 \pi^{2} N}{V}\right)^{2/3}$$

D. The Crystal Lattice

In a crystalline solid the atomic nuclei form a periodic array known as the crystal lattice. Due to the periodicity of this array it is possible to generate the entire set of lattice points by use of the concept of a unit cell. (Ziman, 1964) If the unit cell can be chosen to be a parallelepiped containing one atom, the crystal lattice is said to be a Bravais lattice. On the other hand, if a unit cell with more than one atom is required the crystal lattice is said to be a Bravais lattice with a basis since the positions of the various atoms in the unit cell must be specified. It is easily seen that if the crystal lattice is considered to be made up of unit cells, each containing one atom (Bravais lattice), any lattice point can be reached from any other by a translation through a vector of the form

$$\vec{T} = 1_1 \vec{a}_1 + 1_2 \vec{a}_2 + 1_3 \vec{a}_3$$
 (2-8)

where \vec{a}_1 , \vec{a}_2 , and \vec{a}_3 are vectors defined by the edges of the unit cell, and

or,

 1_1 , 1_2 , and 1_3 are integers. Equation (2-8) is also valid for a crystal lattice made up of unit cells containing more than one atom each (Bravais lattice with a basis). However, in this case the two atoms so linked must reside at equivalent sites within their respective unit cells.

Most metals crystallize in one of three structures. These three are the body-centered cubic, the face-centered cubic and the hexagonal closepacked structures, diagrams of which are shown in Figures 1, 2, and 3, respectively. Nearly all of the common metals have one of these structures. For example, lithium, sodium, and potassium are body-centered cubic whereas copper, silver and gold are face-centered cubic. Examples of the hexagonal close-packed structure are beryllium, magnesium and zinc.







E. The Reciprocal Lattice

In the theory of metals it is convenient to introduce a lattice known as the reciprocal lattice. (Ziman, 1964) This lattice can be defined by means of the reciprocal lattice vector

$$\vec{G} = m_1 \vec{b}_1 + m_2 \vec{b}_2 + m_3 \vec{b}_3$$
 (2-9)

where $\vec{b}_1 = 2\pi \frac{\vec{a}_2 \times \vec{a}_3}{\vec{a}_1 \cdot \vec{a}_2 \times \vec{a}_3}$, $\vec{b}_2 = \frac{2\pi \vec{a}_3 \times \vec{a}_1}{\vec{a}_1 \cdot \vec{a}_2 \times \vec{a}_3}$, $\vec{b}_3 = \frac{2\pi \vec{a}_1 \times \vec{a}_3}{\vec{a}_1 \cdot \vec{a}_2 \times \vec{a}_3}$ and m_1 , m_2 , and m_3

may be positive or negative integers or zero.

The unit cell, or zone, in the reciprocal lattice is then obtained (Kittel, 1956) by finding the shortest non-zero reciprocal lattice vectors satisfying the Bragg condition

$$(\vec{k} + \vec{G})^{L} = k^{2}$$
 (2-10)

where \vec{k} is a general vector in the reciprocal space. From this equation it can be seen that $2\vec{k}\cdot\vec{G} = -G$ so that each zone boundary is normal to a reciprocal lattice vector at its midpoint. The cell thus formed is known as the first Brillouin zone or the reduced zone.

In particular, for a face-centered cubic lattice one may take the primitive translation vectors to be

$$\vec{a}_1 = \frac{a}{2}(\hat{j} + \hat{j})$$

 $\vec{a}_2 = \frac{a}{2}(\hat{j} + \hat{k})$
 $\vec{a}_3 = \frac{a}{2}(\hat{i} + \hat{k})$

where a is the length of a cube edge and \hat{i} , \hat{j} , and \hat{k} are unit vectors along

the cube edges. From the above equations one then easily obtains

$$\hat{s} = \frac{2\pi}{a} \left[(m_1 - m_2 + m_3) \hat{i} + (m_1 + m_2 - m_3) \hat{j} + (-m_1 + m_2 + m_3) \hat{k} \right]$$

Thus the shortest non-zero reciprocal lattice vectors are the eight vectors $\frac{2\pi}{a}(\pm \hat{1}\pm \hat{j}\pm \hat{k})$ and the next shortest are the six vectors $\frac{2\pi}{a}(\pm 2\hat{1})$; $\frac{2\pi}{a}(\pm 2\hat{j})$; $\frac{2\pi}{a}(\pm 2\hat{j})$; $\frac{2\pi}{a}(\pm 2\hat{k})$. The intersection of the planes normal to these vectors (at their midpoints) defines the first Brillouin zone shown as the truncated octahedron in Figure 4.

F. Motion of an Electron in a Crystal Lattice

If the first two terms in the Hamiltonian of (2-1) are retained and the assumption made that the ion cores are closed shells of electrons rigidly attached to their nuclei so that one has a system of non-interacting electrons moving in a lattice of ion cores, equation (2-1) reduces to

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V(F)\right]\psi = \mathcal{E}\psi$$
(2-11)

where V(\vec{r}) is the periodic potential due to the lattice. Now consider a simple Bravais lattice with dimensions N₁a₁, N₂a₂, and N₃a₃ along \vec{a}_1 , \vec{a}_2 , and \vec{a}_3 respectively; where N₁, N₂, and N₃ are integers. The potential energy of an electron in the lattice will have the periodicity of the lattice:

$$V(\vec{r}) = V(\vec{r} + \vec{1})$$
 (2-12)

It is well known (Heine, 1960; Tinkham, 1964) that solutions of (2-11) subject ik \vec{r} to the conditions (2-12) are the Bloch functions $\Psi_{\vec{k}}(\vec{r}) = e = u_{\vec{k}}(\vec{r})$ where $u_{\vec{k}}(\vec{r})$ has the periodicity of the lattice: $u_{\vec{k}}(\vec{r}) = u_{\vec{k}}(\vec{r}+\vec{n})$. Application of the usual periodic boundary conditions $\Psi_{\vec{k}}(\vec{r}) = \Psi_{\vec{k}}(\vec{r}+N_1\vec{a}_1) = \Psi_{\vec{k}}(\vec{r}+N_2\vec{a}_2) =$ $\Psi_{\vec{k}}(\vec{r}+N_3\vec{a}_3)$ gives $\vec{k}\cdot\vec{a}_1 = 2\pi n N_1$, $\vec{k}_2\vec{a}_2 = 2\pi n N_2$, $\vec{k}_3\vec{a}_3 = 2\pi n N_3$ whence $\vec{k} = n N_1\vec{b}_1 + n N_2\vec{b}_2 + n N_3\vec{b}_3$



where n_1 , n_2 , and n_3 are integers. From this expression for \vec{k} it is seen that, just as in the free electron case, there will be $\frac{vd\vec{k}}{8\pi^3}$ orbital states in a volume element $d\vec{k}$ of \vec{k} -space. Use of (2-8) and (2-9) gives e = 1since $\vec{a}_i \cdot \vec{b}_j = 2\pi \delta_{ij}$. Thus it follows that the vector \vec{k} is not uniquely dei($\vec{k} + \vec{G}$) $\vec{r} - i\vec{G} \cdot \vec{r}$ termined since $\Psi_{\vec{k}}(\vec{r}) = e$ [e u (\vec{r})] is also of Bloch form. For this reason the vector \vec{k} is often restricted to the first Brillouin zone (reduced zone scheme). In this scheme the energy is a many-valued function of \vec{k} .

By translating regions of the first Brillouin zone through reciprocal lattice vectors \vec{G} , one can form polyhedra that symmetrically surround the first zone. The faces of these polyhedra are again given by condition (2-10). The volume between the first zone and the next polyhedron defines the second Brillouin zone. Higher zones are defined in a similar manner. By use of this extended zone scheme the energy $\boldsymbol{\xi}$ (k) can be written as a singlevalued function of \vec{k} for all \vec{k} . The extended zone scheme then permits easy comparison of an energy spectrum $\boldsymbol{\xi}$ (k) with the parabolic free electron spectrum $\boldsymbol{\xi}^{\circ}(\mathbf{k}) = \frac{\hbar^2 \mathbf{k}^2}{2m}$.

1. Perturbation Theory for Weak Periodic Potentials

If the periodic potential $V(\vec{r})$ of the crystal lattice is assumed small, standard perturbation theory (Anderson, 1963; Ziman, 1964) yields for the electron energy

where $\mathbf{\epsilon}^{\bullet}(\mathbf{\vec{k}}) = \frac{\hbar^2 k^2}{2m}$ is the energy of the unperturbed free electron state $|\mathbf{\vec{k}}\rangle$. However, since V ($\mathbf{\vec{r}}$) has the periodicity of the lattice it may be written as a Fourier series (Ziman, 1964)

$$V(\vec{r}) = \sum_{\substack{V \\ \underline{G} \\ \underline{G} \\ \underline{G}}} e^{i\underline{G} \cdot \underline{r}}$$

so that the matrix element $\langle \vec{k} | V(\vec{r}) | \vec{k}^1 \rangle$ is nonzero only if $\vec{k} - \vec{k}' + \vec{G} = 0$. If this condition is satisfied, the matrix element is simply $\langle \vec{k} | V(\vec{r}) | \vec{k} \rangle = V_{\underline{G}}$ and the expression for the energy reduces to

$$\mathcal{E}(\vec{k}) = \tilde{\mathcal{E}}(\vec{k}) + v_{o} + \underbrace{\mathcal{E}}_{\vec{G} \neq o} \frac{|v_{G'}|^{2}}{\mathcal{E}(\vec{k}) - \mathcal{E}^{\circ}(\vec{k} + \vec{G})}$$
(2-13)

From this equation it follows that the energy surfaces $\boldsymbol{\epsilon} = \boldsymbol{\epsilon}(\mathbf{\vec{k}})$ will in general not be spherical. In particular, the Fermi surface $\boldsymbol{\epsilon}(\mathbf{\vec{k}}) = \boldsymbol{\epsilon}_{\boldsymbol{F}}$ where $\boldsymbol{\epsilon}_{\boldsymbol{F}}$ is the highest occupied energy level at the absolute zero of temperature, need not be spherical, in contrast to the perfectly free electron case for which $\boldsymbol{\epsilon}_{\boldsymbol{F}} = \frac{\hbar^2 k^2}{2m}$.

From the perturbation expansion (2-13) it can be seen that the method fails if a degeneracy $\vec{c}(\vec{k}) = \vec{c}(\vec{k} + \vec{c})$ occurs. This degeneracy condition is equivalent to the Bragg condition $\vec{k}^2 = (\vec{k} + \vec{c})^2$ of (2-10). In other words, the perturbation expansion (2-13) is not valid when k lies near a Brillouin zone boundary. However, near a zone boundary it is still possible (Ziman, 1964) to expand the electron wave function in a series of the form

$$\Psi_{\vec{k}}(\vec{r}) = e \qquad u_{\vec{k}}(\vec{r}) = e \left(\sum_{\underline{G}} a_{\underline{k}+\underline{G}} e \right) \qquad (2-14)$$

When \vec{k} lies near the zone boundary defined by $\vec{k}^2 = (\vec{k} + \vec{G})^2$ it is possible, as a first approximation, to ignore all coefficients $a_{\underline{k}+\underline{G}}$ except $a_{\underline{k}}$ and $a_{\underline{k}+\underline{G}}$. in order to obtain an expression for the energy. The energy is then given by

$$\mathbf{\mathcal{E}}_{(\vec{k})} = \mathbf{v}_{0} + 1/2 \ \left(\ \mathbf{\mathcal{E}}_{(\vec{k})}^{*} + \ \mathbf{\mathcal{E}}_{(\vec{k}+\vec{g})}^{*} \pm 1/2 \right) \ \left(\ \mathbf{\mathcal{E}}_{\underline{k}}^{*} - \ \mathbf{\mathcal{E}}_{\underline{k}+\underline{G}}^{*} \right)^{2} + 4 \ \left| \ \mathbf{v}_{\underline{G}} \right|^{2}$$
(2-15)

where the minus sign refers to states "inside" the zone boundary ($|\vec{k} \cdot \vec{G}| \cdot |\vec{G}|$)

and where the plus sign refers to states "outside" the zone boundary

 $\left(\begin{array}{ccc} |\vec{k} \cdot \vec{G}| > |\vec{G}| \right)$. From (2-15) it is again seen that in general the Fermi surface will be non-spherical. In addition, the energy spectrum $\mathcal{E} = \mathcal{E}(\vec{k})$ will generally possess discontinuities at the zone boundaries. A detailed discussion of these discontinuities may be found in many textbooks, for example, Kittel (1955), Anderson (1963), or Ziman (1964).

2. Effect of Electron Correlations on the Fermi Surface

Since the concept of a well-defined Fermi surface is based upon the one-electron approximation which largely neglects electron-electron effects, it is of considerable interest to examine the effects of electron-electron interaction on the Fermi surface. Such considerations (Luttinger, 1960; Cornwell, 1964) show that a system of interacting electrons in a crystal does possess a "sharp" Fermi surface (Luttinger, 1960) although in general it will differ in shape (but not in symmetry) from the Fermi surface of a system of non-interacting electrons. (Cornwell, 1964) Such an analysis also shows that the Fermi surface for interacting electrons has the same symmetry as the Fermi surface for electrons interacting through a Hartree-Fock self-consistent field (Cornwell, 1964).

3. Justification of the One-electron Approximation

A justification of the one-electron approximation for a system of interacting electrons is provided by the Bohm-Pines theory of plasma oscillations. In this theory (Raimes, 1957; 1961) a metal is regarded as a plasma composed of a uniform distribution of positive background charge in which the electrons are embedded. This theory shows that plasma oscillations may be associated with a quantum of energy (plasmon) $hw_p > \epsilon_f$ and that plasmons in

a metal at ordinary temperatures will normally be in their ground state and consequently may be often ignored. The theory also shows that the long-range part of the effective Coulomb interaction is associated with the plasmons. The remaining Coulomb interaction has an effective range of ~ 1 Å which is so short that often it too may be neglected. The experimental work on plasma oscillations has been discussed by Pines (1955, 1956).

4. Energy Band Calculations by use of the One-electron Model

Energy band calculations are in principle based on the numerical solution of the Hartree-Fock equations (Callaway, 1958). As these calculations are difficult various approaches and approximations have been employed. Since the first calculation for sodium by Wigner and Seitz (1933), well over a hundred energy band calculations for metals alone have been published. Many of these calculations have been for the same metal. For example, the calculation for sodium has been made more than ten times by use of various approaches (Slater, 1963).

The agreement of energy band calculations with experiment is often not satisfactory. For the alkali metals the agreement with the available experimental data is fairly satisfactory (Callaway, 1958). On the other hand, with the exception of copper, few detailed calculations for the noble metals have been published. For copper, however, at least a dozen calculations have been performed (Slater, 1963). Agreement of the recent work by Segall (1962) and Burdick (1963) with experiment is good. For the few divalent metals for which detailed band calculations exist, it appears that only for beryllium is there satisfactory agreement between theory and experiment (Callaway, 1958). Relatively few calculations have been performed for trivalent, quadrivalent or pentavalent metals. However, the calculations for aluminum appear to be in qualitative accord with experiment (Callaway, 1958). For the transition elements the calculations are very difficult and the results tend to be rather qualitative (Callaway, 1958).

5. The Fermi Surface of Copper: Theory and Experiment

The energy band calculations of Segall (1962) and Burdick (1963) for copper show quantitative agreement with experiment. For example, Segall {111} directions has calculated the radius of the "necks" which lie in the as well as the average "belly" radius of the Fermi surface. For the neck radius Segall obtains a value of 0.28 ± 0.03 x 10 which compares well cm with the experimental value of 0.26 \times 10 cm (Joseph and Thorsen, 1964). For average "belly" radius he obtains 1.33 ± 0.01 x 10 cm which is in good -1 accord with the experimental value of about 1.32 x 10 cm obtained from the work of Morse (1960). The detailed results of Burdick and Segall are compared with experiment in Table I. The symbols used in Table I are defined in Figure 5. All dimensions are expressed in terms of the free-electron Fermi -1 sphere radius $k_f = 1.365 \times 10 \text{ cm}$. In the table the number (1) under k_f refers to the neck radius as measured along a line passing through the center of the hexagonal zone face and the midpoint of one edge, whereas (2) under k_{r} refers to the neck radius as measured along a line passing through the center of the hexagonal Brillouin zone face and one corner. The numbers (1) to (4)appearing under k_{100} and k_{110} refer to the different values obtained when the direction of the sound waves used in the magnetoacoustic measurements is changed. Further details may be found in the paper by Bohm and Easterling (1962).



COMPARISON OF THEORETICAL AND EXPERIMENTAL FERMI SURFACE DIMENSIONS

FOR COPPER (AFTER BOHM AND EASTERLING, 1962)

FERMI SURFACE BOHM-EASTERLING (a) DIMENSION ROAF (b) k₁₀₀ 1.036 ± 0.021 1.076 (1)(2) 1.100 ± 0.060 ^k110 (1) 0.957 ± 0.011 0.943 (2) 0.956 ± 0.011 (3) 0.959 ± 0.032 (4) 0.956 ± 0.011 0.852 ± 0.009 k k k k k k 0.815 ± 0.021

0.284 ± 0.042

0.195 t 0.011

0.191 ± 0.011

SEGALL (d)

1.02 ± 0.015

0.94 ± 0.015

0.015

0.015

0.02

0.02

1.11

0.87 ±

0.81 ±

0.24 t

0.21 ±

1.09

(a) Magnetoacoustic effect

(1)

(2)

(1)

(2)

 k_{100}/k_{110}

k100/k110

k 100 k 110 k

k^d

kⁿ

- (b) Dimensions deduced by Roaf (1962) from Shoenberg's de Haas-van Alphen data (1962), Pippard's (1957), and Morton's (1960) anomalous skin effect measurements.
- (c) Band theory calculation (Chodorow potential)

1.10

(d) Band theory calculation (\mathcal{L} -dependent potential)

SEGALL (c)

1.04 ± 0.015

0.94 ± 0.015

0.87 ± 0.015

0.76 ± 0.015

 0.29 ± 0.02

 0.14 ± 0.02

(e) Band theory calculation (Chodorow potential)

0.200

1.14

BURDICK (e)

 1.05 ± 0.02

0.97 ± 0.02

0.84 ± 0.02

 0.31 ± 0.04

 0.15 ± 0.04

0.17 ± 0.02

1.09

0.77 ± 0.015

CHAPTER III

ANNIHILATION OF POSITRONS

A. Introduction

In recent years considerable theoretical and experimental work has been done in the field of positron annihilation. This chapter outlines some of the theoretical and experimental results relevant to the study of Fermi surfaces by positron annihilation.

B. Free Annihilation of Positrons

Free positron-electron annihilation may proceed by 1, 2, 3 or more photons, as long as linear momentum, angular momentum, energy and other selection rules are satisfied (Jauch and Rohrlich, 1955). In this respect, one-photon annihilation can only occur in the presence of an external system able to take up the recoil momentum. The one-photon process is rather unlikely and in a typical solid over 95% of the positrons annihilate by two
or more photons.

For positron-electron pairs, aside from selection rule considerations, the probability for annihilation into n+1 photons is smaller than that for annihilation into n photons by a factor of order \propto where \propto is the fine structure constant and n \geq 2. Thus little error will be incurred if processes for which n > 3 are ignored. In fact detailed considerations indicate that the ratio of the spin-averaged cross sections for the two-photon and three-photon processes is $\sigma_{12}/\sigma_{33} = 372$ (Berko and Hereford, 1956). This ratio is in reasonable agreement with the experimental result of Basson (1954) for aluminium, viz. $\sigma_{2}/\sigma_{33} = 406 \pm 50$. Thus, the annihilation of positrons with conduction electrons in a metal occurs predominantly by the emission of two photons (Graham and Stewart, 1954).

The cross section for two-photon annihilation of a free positron with a free electron was first obtained by Dirac (1930) by means of a "plane wave" calculation (implying no Coulomb distortion). In the non-relativistic limit, this result for the spin-averaged cross section per electron reduces to

$$\sigma_{2x} = \pi r_o^2 c / v$$

where $r_{o} = \frac{e_{2}^{2}}{mc^{2}}$ is the classical electron radius and v the relative velocity of the positron and electron. Although this cross section diverges as the relative velocity v approaches zero, the annihilation rate (i.e.probability per unit time of annihilation) of a positron in an electron gas is independent of velocity, being given by

$$R = N v_{2\mathfrak{F}}^{\sigma} = \pi r_{\mathfrak{o}}^{2} cN \qquad (3-1)$$

where N is the electron density.

Because of the success of the electron gas model of the conduction electrons in metals, it might be expected that the mean lifetimes of positrons in metals would be approximately those expected on the basis of (3-1) -9 yielding values of $\Upsilon = \frac{1}{R} \sim 10$ sec and inversely proportional to the conduction electron density. The observed lifetimes, however, are surprisingly -10 constant ($\Upsilon \sim 2 \ge 10$ sec) despite the large variation in electron density from one metal to another (Wallace, 1960). Much of this disagreement probably arises from the neglect of the positron-electron interaction involved in deriving (3-1) and will be examined in the discussion of lifetimes below (Section H).

C. Positron Annihilation from a Bound State

1. Bound Electron-Positron Systems

In addition to these polyelectron systems, dynamically stable bound states of positrons with molecules or ions are also possible. For example, calculations indicate that positron hydride e^+ H⁻ should be stable by about 0.23 ev (Neamtan et al., 1962) and positron chloride by about 4.65 ev (Simons, 1953). Again, as in the case of the three and four-electron systems, little experimental work has been done on these "compounds" (Green and Lee, 1964).

2. Positronium

Positronium bears a resemblance to the hydrogen atom in that it is also composed of a pair of oppositely charged particles. Since the reduced mass of positronium is half that of the hydrogen atom, the positronium energy levels will be half those of hydrogen and its Bohr radius twice as large. Thus the positronium ionization energy is 6.8 ev and the positron Bohr radius is 1.06 Å. A discussion of the fine structure of positronium may be found in, for example, Deutsch (1953).

3. Positronium Annihilation

Positronium decays by two and three quantum annihilation; the ${}^{1}S_{0}$ (para-) state decaying by two-photon annihilation and the ${}^{3}S_{1}$ (ortho-) state by three photons, two-photon decay of the ${}^{3}S_{1}$ state being forbidden by the selection rules. The lifetime for two-photon annihilation is given by $\Upsilon_{1} = 1.25 \times 10^{10} \text{ n}^{3}$ sec and that for three-photon annihilation by $\Upsilon_{3} = 1.4 \times 10^{7} \text{ n}^{3}$ sec. Thus for annihilation from the ground state (n = 1) the lifetimes become $\Upsilon_{1}=1.25 \times 10^{10} \text{ sec}$ and $\Upsilon_{3}=1.4 \times 10^{7} \text{ sec}$. If the positronium is formed in any excited state (other than ${}^{1}S_{0}$ states or the 2 ${}^{3}S_{1}$ state) it should radiate optically to the ground state before annihilating (Deutsch, 1953).

Since positronium formation does not seem likely in metals

(Wallace 1960; Kanazawa et al., 1965; see also, for example, the recent work on lifetimes by Kugel et al., 1966, positronium will not be discussed further in the present work. The rather extensive subject of positronium formation and decay in solids, liquids, and gases has been reviewed by Wallace (1960). Positronium chemistry has been discussed by Green and Lee (1964).

D. Angular Correlation of Two-Photon Annihilation of Positrons

Energy and momentum are conserved if the two photons (each of energy m_c^2) are emitted in opposite directions in the center-of-mass frame (Heitler, 1954). If the center of mass frame is in motion relative to the laboratory reference frame, the angle between the photon directions may depart from 180° . The departure from collinearity of the two photons can be found by transferring the collinear two-photon system from the center of mass frame to the laboratory frame by means of an appropriate Lorentz transformation. The calculation is similar to that used to calculate the aberration of light in relativistic optics (Becker and Sauter, 1964).

If the relative electron velocity v is very small compared to the velocity of light ($v/c \sim 10^3$) it is possible to derive a simple expression for the deviation of the photon directions from antiparallelism. In this case the kinetic energy of the pair is very small compared to the energy carried off by the two photons so that each photon will have an energy very nearly equal to mc². Thus each photon has momentum mc, and since is small it is seen from Figure 6 that the transverse component of pair momentum is given by

$$l_{t} \approx m_{o} c \Theta$$

(3-2)

This is the fundamental angle-momentum relation used in two-photon angular correlation studies.



E. Angular Correlation Geometries

1. Introduction

There are essentially three ways by which the Fermi surface has been studied by use of angular correlation of annihilation radiation. Most angular correlation experiments have used the "wide slit" geometry described below (Stewart, 1957) although some recent work has been done by use of a "point" geometry (Colombino et al., 1963, 1964' Fujiwara 1965). The work described in this thesis utilizes a third method ("collinear point geometry") in which the angle does not vary. These methods are outlined below.

2 (a) Wide Slit Geometry

There are several variations of the wide slit method (Stewart, 1957; Berko and Plaskett, 1958) but it is sufficient to consider Stewart's arrangement since the methods are equivalent. In this arrangement, shown in Figure 7, the gamma-ray pairs are counted in time coincidence by means of detectors mounted behind two slits that can be taken to lie in the horizontal plane. The radioactive sample containing annihilating electron-positron pairs is allowed to move in a line perpendicular to the plane of, and midway between, these two slits. By moving the source vertically, photons emitted in time coincidence at various angles may be observed. Some typical dimensions (Stewart, 1957) for the distances shown in the figure are

> D = 100 in. l = 1.5 in. h = 0.050 in.

Further experimental details can be found in Stewart's paper.

From Figure 7 and by (3-2) it is seen that, approximately,

$$p_{r} = mc\theta = 2mcz/D \qquad (3-3)$$

where z is the vertical displacement. Thus in this method the transverse component of pair momentum p_{r} is directly proportional to the displacement z.

If now this approach is considered in terms of the free-electron model it is seen that for each setting z of the displacement, one samples a slice through the Fermi volume as shown in Figure 9. The slice thickness, of course, is determined by the finite resolution of the experimental arrangement.

Following the analysis of Stewart (1957) it is seen that the coincidence count rate will be given by

$$n(p_{z}) \propto \int_{-\infty} \int_{-\infty} f(\vec{p}) dp_{x} dp_{y}$$

where $\beta(\vec{p})$ is the density of momenta of the annihilating pairs. If $\beta(\vec{p})$ is assumed isotropic so that $\beta(\vec{p}) = \beta(p)$, the count rate becomes

$$n(p_z) \propto \int_{0}^{\infty} \rho(p) 2\pi p_p dp = 2\pi \int_{\rho_z}^{\infty} \rho(p) p dp$$

where $p^2 = p_r^2 + p_z^2 = p_x^2 + p_y^2 + p_z^2$. Thus, differentiation of this expression for $n(p_z)$ gives

$$\int (p_{\mathbf{x}}) \propto - \frac{1}{p_{\mathbf{z}}} \frac{dn(p_{\mathbf{x}})}{dp_{\mathbf{z}}}$$

so that by (3-3) it follows that

$$f(\mathbf{p}_{\mathbf{z}}) \propto -\frac{1}{z} \quad \frac{\mathrm{dn}(z)}{\mathrm{dz}}.$$
(3-4)

For the ideal free-electron case in which it is assumed that:



- (a) The momentum of the positron is zero (i.e. the momenta of the thermalized positrons are neglected compared to the electron momenta.
- (b) The electron momentum distribution is isotropic and taken to be of uniform density $\int p$ up to the maximum, or Fermi momentum p and zero for p > p.

one has

$$n(p_z) \ll \int_{r_z}^{r_F} pdp$$

or

$$n(p_{z}) \propto (p_{z}^{2} - p_{z}^{2})$$
 (3-5)

which is the equation of an inverted parabola.

As might be expected, the experimental counting rate curves deviate from an exact inverted parabola. In the case of the alkali metals the deviation is small. This confirms the statement that the alkali metals exhibit very nearly free electron behaviour. But as one passes through the alkaline earths and beyond, increasingly larger "tails" are superimposed on the inverted parabola. For comparison, the results for sodium and copper (Stewart, 1957) are shown in Figure 8. In this figure O denotes the angle corresponding to the Fermi momentum p_{p} . Berko and Plaskett attribute most of the tail of the copper curve to annihilation of positrons with the electrons of the ion cores (core annihilation). They have estimated the core contribution for copper and find that their estimate is in reasonable agreement with experiment. However, it should be noted that there are difficulties associated with these core annihilation calculations. These difficulties will be discussed below.

2 (b) Determination of Fermi Surfaces by Wide Slit Geometry

If a particular direction of a metal single crystal is aligned with the z axis in the wide slit method, it is found that the shape of the angular correlation curve may depend upon the choice of crystal orientation (Berko and Plaskett, 1958). Although this anisotropy has been interpreted as being due to a nonspherical Fermi surface it is usually difficult to make quantitative statements about the topology of the Fermi surface on the basis of such measurements. A major source of difficulty is the fact that the method samples a slice through momentum space, giving a result that is an average over a considerable region. In addition, this averaging makes it difficult to observe the sharp "break" in the angular correlation curve which should (Majumdar, 1965) occur at angles corresponding to the Fermi momentum. Also, recent work with lithium by Donaghy et al. (1965) suggests that the higher momentum components of the electron wave function may modify the shape of the angular correlation curve sufficiently so that the observed "breaks" in the angular correlation curve no longer correspond to radii (Stewart et al., 1962) of the Fermi surface. Despite these limitations, the method has been used to study the Fermi surface of a number of metals; in several cases confirming qualitatively the results obtained by the more precise conventional methods. Metals in the form of single crystals that have been examined by this method include sodium (Donaghy et al., 1965), beryllium (Stewart et al., 1962; Berko, 1962), magnesium (Berko, 1962), aluminum and copper (Berko and Plaskett, 1958) as well as holmium, erbium, and yttrium (Williams et al., 1966).

3 (a) Point Geometry

In this method two "point" detectors are used in place of the wide slit arrangement discussed above. With this arrangement the region of momen-

tum space that is sampled is an infinite cylinder whose diameter is determined by the detector geometry. The arrangement is illustrated in Figure 10.

Using a notation similar to that used in the discussion of the wide slit case, it is seen that for counting rate at p_z one expects (Figure 11)

$$\pi(p_{z}) \propto \int_{0}^{\infty} g(p) \, dp_{p} = \int_{0}^{\infty} \frac{g(p) \, p \, dp}{\sqrt{p^{2} - p_{z}^{2}}} \tag{3-6}$$

The solution of this integral equation (Appendix I) is

$$\int (p_2) \propto -\frac{1}{P_z} \frac{d}{dp_z} \int_{\frac{P}{p_z}}^{\infty} \frac{\frac{P n(p)dp}{p_z}}{\sqrt{p^2 - p_z^2}}$$
(3-8)

If, as for the wide slit case, f(p) is taken to be constant inside the Fermi surface and zero outside, it is seen from (3-6) that the counting rate is given by

$$n(p_z) \ll \sqrt{p_f^2 - p_z^2}$$
(3-9)

so that a plot of n^2 vs. p_z^2 is linear.

3 (b) <u>Determination of Fermi Surfaces by Use of Point Geometry</u>

The use of point geometry would be expected to yield angular correlation curves containing more structure than those characterizing the wide slit geometry since the method samples a smaller region in momentum space. A preliminary study of the Fermi surface of copper has been made by this method (Fujiwara, 1965). Although the results definitely show more detail than do those of Berko and Plaskett (1958), the copious presence of higher momentum components again makes quantitative interpretation in terms of the Fermi surface difficult.



4. Collinear Point Geometry

In this method the radioactive metal single crystal and the two "point" detectors remain collinear and the coincidence count rate obtained as a function of crystal orientation (Figure 12). On the basis of the freeelectron model this method also samples a cylindrical volume of momentum space. This volume element rotates about the origin of momentum space as the crystal orientation varies so that one would expect variations in counting rate with crystal orientation to be interpretable in terms of radii to the Fermi surface. That is, from (3-9), if Θ (and hence p_z) is zero, then n $\propto p_f$. If it is assumed that contributions to the counting rate from core annihilations are isotropic and if it is assumed that the net effect of all other higher momentum effects is roughly isotropic, then variations in the coincidence counting rate are a direct measure of fluctuations of the Fermi surface radius.

F. Annihilation of Positrons in Electron Gases

When positrons annihilate in a dense gas of interacting electrons the momentum distribution of the photon pairs should differ from the simple Fermi-Dirac momentum distribution characteristic of non-interacting electrons and shown as curve a of Figure 13. This deviation arises from the effect of electron-electron and electron-positron correlations (Daniel and Vosko,1960; Kahana, 1960; 1963). Several attempts have been made to calculate the effect of these various correlations on the momentum distribution of the gamma-ray pairs (Hatano et al., 1965).

1. Effect of Electron-electron Interactions

If the electron-positron correlations are ignored and only the

electron-electron correlations taken into consideration, the momentum distribution of the gamma-ray pairs will be identical to that of the interacting electrons (Hatano et al., 1965). Daniel and Vosko have calculated the momentum distribution of such a system of interacting electrons, and for an electronic density corresponding to that in sodium they obtain the results shown in curve b of Figure 13. It is to be noted that in this case the positron acts as an ideal probe since electron-positron correlations have been ignored.

2. Effect of Electron-Positron Interactions

By an approximate solution of a Bethe-Goldstone equation for a positron-electron pair in a sea of interacting electrons, Kahana (1960;1963) obtained the photon pair momentum distribution given by curve c of Figure 13. In this calculation the two-body correlations between the positron and the annihilating electron have been accurately accounted for. However, with the exception of the correlations associated with the screening of the attractive electron-positron force, no other correlations are considered. Hatano et al. (1965) have suggested that the neglect of these other correlations may be justified only for high electron densities and not for the relatively low electron densities found in real metals.

3. Effect of Electron-electron and Electron-positron Interactions

By starting with a wave function of Bijl-Dingle-Jastrow type, Hatano et al. have been able to approximately include both electron-electron and electron-positron correlations. Their result is shown in curve d of Figure 13. For the relative momentum distribution of the photon pairs they obtain

$$\frac{w(p)}{w(0)} = 1 + 0.13 (p/p_f)^2 - 0.02 (p/p_f)^4 + \dots$$

in which the coefficients are independent of electron density. This is to be compared with Kahana's result(for an electron density corresponding roughly to sodium)

$$\frac{w(p)}{w(0)} = 1 + 0.262(p/p_f)^2 + 0.233(p/p_f)^4$$

where $p_f = \hbar k_f$ is the usual Fermi momentum. It is seen that there is a substantial difference between the two results.

4. Comparison with Experiment

Since the conduction electrons in sodium can be considered to roughly approximate an electron gas it might be reasonable to assume that experimental studies of sodium could be used to choose the "best" of the theoretical curves shown in Figure 13 and thus give some insight into the annihilation process. However, this is rather difficult to do because of experimental uncertainties. For example, although the computations by Hatano et al. appear to be in better agreement with Stewart's experimental results for sodium than are those of Kahana or Daniel and Vosko, Stewart's results (1961) are explained just as satisfactorily by the simple Sommerfeld model (curve a, Figure 13). On the other hand, recent work by Donaghy et al. (1965) with sodium appears to give results that are in good accord with Kahana's curve shown in Figure 13. It is important to note however, that the results of Donaghy et al. involve a correction for core annihilation (to be discussed below) which is rather difficult to estimate (Carbotte, 1966). Thus on the basis of angular correlation work alone it is difficult at present to make a definite choice between the various models.



G. Annihilation of Positrons in Real Metals

1. Introduction

A quantitative treatment of the effect of the crystal lattice upon the angular correlation of annihilation radiation from metals is exceedingly difficult (Wallace, 1960; Carbotte, 1966). One of the sources of difficulty is the presence of higher momentum components in both the positron and electron wave functions due to the periodic potential of the crystal lattice. Another complication arises from the annihilation of positrons with core electrons. Both of these effects yield a contribution to the angular correlation at values of **9** greater than those which would arise from annihilation with free conduction electrons alone.

2. Effect of the Periodic Crystal Lattice Potential

An early attempt to consider the effect of the crystal lattice was made by De Benedetti et al. (1950). They considered a simple model in which, due to Coulomb repulsion, the positron wave function is excluded from a volume v_e (the "excluded volume") around the nucleus. Outside this excluded volume the positron wave function is taken to be a constant and the electron wave function is considered a simple plane wave. The restriction of the positron to regions surrounding the "excluded volume" regions thereby introduces higher momentum components into the positron wave function as would be expected, for example, from the Heisenberg uncertainty principle. The relative intensity of these higher momentum components is related to the quantity v_e/v which is the ratio of the excluded volume to the volume of a unit cell. Typical values of v_e/v range from 0.05 in beryllium to 0.29 in barium (Lang, 1956; Lang and De Benedetti, 1957). The results however generally disagree

with experiment (Lang, 1956; Berko and Plaskett, 1958), the disagreement probably being due to core annihilation and to contributions from the electron positron interaction (Wallace, 1960).

3. Effect of Core Annihilations

By use of uncorrelated one-particle electron wave functions Berko and Plaskett (1958) have calculated the angular distribution of photons resulting from the annihilation of positrons with core electrons. The calculations were done for copper and aluminum and are in fairly good agreement with experiment. However, the computed momentum distribution is quite sensitive to the shape of the positron wave function so that it is difficult to assess the validity of the method. Also, recent calculations by Carbotte (1966) indicate that the electron-positron interaction may be of considerable importance in core annihilation calculations. The Berko-Plaskett method has been applied to other metals by Rockmore and Stewart (1965) and by Terrell et al. (1965).

H. Lifetimes of Positrons in Metals

If typical values of the electron density of metals are substituted into (3-1), lifetimes of the order of 10^9 sec are obtained. This is in disagreement with the experimentally observed lifetimes of $\sim 2 \times 10^{10}$ sec. In addition, the observed lifetimes are essentially independent of electron density, (Wallace, 1960) whereas (3-1) predicts a direct dependence on the electron density. Thus annihilation in metals cannot be regarded as free annihilation in which the effects of the Coulomb forces can be neglected. Neither can the lifetimes be interpreted in terms of positronium formation since triplet positronium would have a long lifetime of about 10^7 sec which

is not observed. On the other hand, rapid triplet to singlet conversion would lead to a lifetime of ~5 x 10^{10} sec, four times that of singlet positronium (Wallace, 1960). However, this is also considerably larger than the observed lifetimes of ~2 x 10^{10} sec.

The effects of electron-positron correlations on the annihilation rate in metals have been investigated by Ferrell (1956), Kahana (1963), Carbotte and Kahana (1965), and Kanazawa et al. (1965). These studies indicate that the positron lifetime is very sensitive to correlation effects, the annihilation rate being directly dependent on the probability that the positron and electron are at the same place. The recent work of Kahana (1963), which accurately takes into account the two-body correlations between the positron and the annihilating electron, predicts positron lifetimes that are in order of magnitude agreement with experiment (Figure 14). His work also gives an angular correlation curve that is in rough agreement with the experimental work of Stewart (1961). Kanazawa et al. (1965) have calculated the annihilation rate for electrons of zero momentum, their results being shown in Figure 14. Inclusion of effects of the crystal lattice into lifetime calculations is also very difficult although preliminary work by Carbotte (1966) has already provided some insight into the core annihilation process.

.

CHAPTER IV

EXPERIMENTAL ARRANGEMENT

The schematic diagram in Figure 15 shows the experimental arrangement for the "collinear point geometry" method used in the present work. In order to be detected, photons from the positron-active single crystal of copper had to pass through the 0.25 inch diameter aperture of the lead collimators that were placed at a distance of about 12 feet from the copper crystal. If a gamma-ray pair was detected by the two counters within the coincidence circuit resolving time of 10 nsec, a coincidence was recorded by the scaler. In this way, within the resolution of the apparatus, only photon pairs of zero transverse momentum were counted. For each orientation of the crystal the time interval required for recording a predetermined number of counts was measured. This made it possible to accurately correct for the decay of the positron activity ($T_1 = 12.9$ hr) and thus obtain a measure of momentum space anisotropy as discussed in Chapter III.



A. Metal Crystal and Positron Source

The copper crystal used in this experiment was cylindrical, 3.56 mm in diameter by 3.84 mm high, with [111] direction along its axis. Copper was chosen as the metal to be examined by this method for several reasons:

- The topology of the Fermi surface of copper is well known (Chapter II) thus making it relatively easy to ascertain the usefulness of the new positron annihilation technique used in this experiment.
- Copper is stable at ordinary temperatures and is readily obtained in the form of very pure single crystals so that sample environment or sample impurities need not be considered.
- 3. The use of a conventional positron source was rendered unnecessary since by thermal neutron irradiation the copper crystal itself became an adequate positron source.

The present approach was advantageous since it provided a nearly uniform distribution of positrons throughout the crystal whereas conventional source geometries "shine" positrons on the sample surface (Berko and Plaskett, 1958). Also, in contrast to some positron sources, the gamma radiation emanating from the crystal is nearly all annihilation radiation thus ensuring noninterference from other radiations. Another important consideration is that of economy. A conventional long-lived source of comparable strength would have been prohibitively expensive.

The main disadvantage of this type of positron source is the inconvenience associated with the short half-life. Due to the relatively low coincidence counting rates resulting from use of "point" geometry it was

necessary to have a relatively high initial positron activity (~50 mCi). Although the coincidence counting rate at the beginning of a run was adequate (~1 count per second) this rate falls with time, making it difficult to establish many experimental points with good statistics. The use of a longlived source would have obviated this difficulty.

The size of the crystal was determined after consideration of several conflicting requirements. From the point of view of angular resolution, the ideal crystal should be vanishingly small. On the other hand, it is necessary to have the crystal larger than a certain minimum size in order that it have sufficient positron activity upon irradiation with the thermal neutron fluxes currently available. In addition, it was desirable to choose the crystal large enough so that few of the positrons produced in the crystal escape. Taking these considerations into account, the sample was chosen to be a cylinder with diameter roughly equal to height so that its extent, as seen from the "point" detectors, would be as small as possible. With the dimensions given above it is easy to estimate, by use of results from Price et al. (1957) and Evans (1955), that less than five percent of the positrons will escape from the sample.

B. Orientation of Crystal

The copper crystal used in the present work was a cylinder possessing a [111] direction along its axis. By means of x-ray diffraction (Laue back-reflection) the other $\{111\}$ directions of the crystal were determined relative to a fiducial line on the crystal base. The crystal was then placed in a "notch and pin" arrangement (Figure 16) which, upon rotation, brought successive $\{111\}$ directions of the crystal into the direction defined by the two "point" detectors. Thus, as can be seen from the discussion



in Chapters II and III, the necks of the Fermi surface (which for copper occur in the $\{111\}$ directions) can be examined by this arrangement.

C. Spatial Stability of Notch and Pin Assembly

In the design and construction of the notch and pin assembly considerable care was taken to maximize the spatial stability of the arrangement. Stability measurements were made by varying the vernier dial setting and carefully observing the crystal position. The measurements indicated that motion of the centre of mass of the crystal from its mean position was less than 0.10 inches along the direction (vertical) of the detectors and less than 0.004 inches horizontally. This residual mechanical instability causes an uncertainty of \sim 0.2 percent in the coincidence count-rate. As will be seen from the results presented in Chapter V, this uncertainty is small compared to the uncertainty engendered by counting statistics or electronic drift.

D. Crystal Holder

In the first run the copper crystal was attached by means of epoxy adhesive to a small cylindrical holder made of high purity aluminum. High purity aluminum was used for the holder since aluminum has a small neutron absorption cross section and in addition, the activity induced in irradiated aluminum has a very short half-life (2.30 min.). The cylinder composed of crystal and holder was designed to fit into a tube containing a notch which engaged a small pin on the holder (Figure 16).

This method was used for the first experimental run, but contrary to expectation (Livingstone, 1963) the epoxy suffered considerable radiation damage so that upon arrival the crystal was found separated from the holder. This made it necessary to remount the copper crystal on a new cylinder by use of fresh epoxy before a run could be made.

In the second run the crystal was force fitted (before irradiation) into the end of a thin-walled aluminum holder as indicated in Figure 17. This holder had about the same gross dimensions (4.72 mm diameter by 39.4 mm long) as did the first except that it had a hollow central region to further reduce scattering of the annihilation radiation. With this arrangement there is little attenuation of the gamma radiation by the thin covering (0.033 inches) of aluminum.

E. Holder Support

A simplified schematic diagram of the holder support is shown in Figure 16. The crystal holder fits into the supporting tube which contains a notch that engages the pin on the crystal holder. This tube was attached to a vernier dial (Armaco DV4) that was securely bolted to a heavy aluminum plate. This heavy plate in turn was bolted to an aluminum beam that was cantilevered at its other end. The supporting tube was stabilized by means of a system of discs and rods which, for clarity, have been omitted from the figure. The use of these stabilizing elements made it possible to reduce the residual mechanical instability of the crystal to the negligible amount mentioned in the stability discussion above.

To the end of the vernier shaft was attached a simple pulley wheel which could accommodate a small diameter cord. By use of the cord and another pulley wheel attached to a long, supported rod it was possible to easily control the vernier dial setting from a distance of about ten feet from the radioactive copper crystal. (Figure 18) The vernier dial was conveniently



read from this distance by means of a telescope mounted on a tripod. With this arrangement the radiation hazard to the operator was found to be small (less than 5 mrem per 8 hours).

F. Gamma Counters

The "point" detectors consisted of two Harshaw NAI(TI) crystals mounted on RCA 6342 photomultiplier tubes. These crystals were cylinders one inch in diameter by two inches long. The 6342 photomultiplier tube was chosen because it was economical and readily available. The 6342 also has a small transit time spread (~4 nsec), thus facilitating the short coincidence resolution time. On the other hand, the detector crystals were chosen as a compromise between economy and efficiency. For example, the measured efficiency of the individual detectors was about 0.45, to double this would probably require a crystal at least four inches in diameter and four inches in height.

The detector assemblies were enclosed in light-tight cylindrical aluminum housings, each photomultiplier tube being magnetically shielded by means of a "Co-netic Netic" alloy shield. (manufactured by Perfection Mica Co.) In addition each aluminum housing was wrapped with about 0.2 inches of lead foil for radiation shielding purposes. Each detector was rigidly bolted to a large (6x6x3 in) lead block containing a cylindrical aperture 0.25 inches in diameter and 3 inches long. The source to detector distance was twelve feet.

The choice of source to detector distance and collimator aperture size was based on several considerations. These will now be briefly dis-cussed.

The most important consideration was that of resolution. From Table I it can be seen that the diameter of the Fermi surface "necks" is $2r_{o} \approx 10^{-3}$ mc. Hence, in order to observe a reasonable coincidence counting rate change (about half that expected for a point source and point detectors) at crystal orientations associated with the necks, a resolution function (Chapter V) with full width at half maximum of $W_{1/2} \approx 4r_{o}$ is required. This choice of $W_{1/2}$ will be further discussed below.

In the present work, a choice of D = 12 feet for the source to detector distance was particularly convenient since this was the vertical distance between successive floors of the tower which housed the experimental arrangement. Since the condition $2D > c \tau$, where τ is the coincidence circuit resolving time, is amply satisfied it is seen that the cosmic ray contribution to the coincidence count rate should be negligible. Indeed, an experimental check (Chapter V) showed that the total background co-incidence rate (i.e. the rate with the radioactive copper crystal absent) was also very small. For D = 12 feet a choice of d = 0.25 inches for the collimator aperture diameter then gives (Chapter V) a resolution function halfwidth of $W_{1/2}^{\infty} 4r_0$ as required.

The choice of $W_{1/2}^{\sim} 4r_o$ was a compromise between coincidence count rate and sensitivity to the Fermi surface topology. For example, reduction of the resolution function width from $W_{1/2}^{\sim} 4r_o$ to $W_{1/2}^{\sim} 2r_o$ would reduce the coincidence count rate by a factor ~16 (from about 60 counts per minute (maximum) to about 4 counts per minute (maximum).) Problems associated with the specific activity of the source must also be taken into consideration. These problems are further discussed in Chapter V.

G. <u>Electronics</u>

The schematic diagram of Figure 15 indicates the electronics used in the experiment. A photomultiplier current pulse arising from a gamma-ray impinging on one of the Nal(Tl) crystals is amplified, discriminated, and shaped, giving as output a positive pulse about 25 nsec wide. Such pulses were fed into a coincidence unit with resolving time set at ~10 nsec. The resulting coincidences were then recorded by a E 110A decade scaler. The E 110A was manufactured by Oxford Engineering Corp. (this firm is no longer in existence).

The circuit diagrams of the preamplifier and shaping circuit and of the coincidence circuit are shown in Figures 19 and 20 respectively. After differentiation and preamplification, the negative current pulse from the photomultiplier collector was clipped by a shorted delay line, giving a bipolar zero-crossing signal. This signal, when of sufficient amplitude, triggered a Schmitt-type, zero-crossover discriminator circuit. The output from the discriminator was then shaped to give a 25 nsec output pulse which went into the coincidence circuit shown in Figure 20. In this circuit adjustment of the coincidence sensitivity control varies the input threshold level. For the present work this control was adjusted to give a standing current of about 1 ma through the tunnel diode. This current was small enough to ensure that an output pulse (- 10 volts) occurred only when two 25 nsec pulses from the pulse shapers arrived simultaneously (ie. within the resolving time of 10 nsec) at the coincidence circuit input.

H. <u>Stability of Electronics</u>

Since the count-rate variation expected from the present method is fairly small (about six percent) it is important that drifts in the coincidence





counting rate be minimized. Several precautions were taken to ensure that large drifts in the coincidence count rate did not occur. One such precaution involved minimizing the fluctuations in the ambient temperature of the tower which housed the apparatus. This was easily done by opening doors in the experimental area so free circulation of air from the main building to the tower was possible. The ambient temperature was monitored throughout the course of the two experiments, the maximum fluctuations from the mean ambient temperature being about $\pm 0.8^{\circ}$ C. During both runs, certain points of the experimental curves (Chapter V) were repeated to ensure that large drifts in the set-up did not occur. To within the statistical uncertainty, the points were found to be reproducible. Tests with a Na²² source indicated that the coincidence counting rate was stable to better than ± 0.6 percent over a 24 hour period.

CHAPTER V

RESULTS AND CONCLUSIONS

A. Introduction

The experimental arrangement is as shown in Figure 15 of Chapter IV. The crystal support was inclined at an angle, as shown, so that the [111] axis was the axis of rotation and so that upon rotation, $\{111\}$ directions would successively point in the direction of the detectors. These $\{111\}$ directions of the copper crystal had previously been determined by standard x-ray techniques at the Department of Metallurgy x-ray facilities.

As pointed out in Chapter III, one would expect the coincidence count rate to rise at crystal orientations corresponding to the "necks" of the Fermi surface, which for copper occur in the $\{111\}$ directions. Also, as a consequence of the three-fold symmetry about a $\{111\}$ axis one would expect peaks in the coincidence count rate at intervals of 120° . From the available experimental data on the Fermi surface of copper (Table I) one would expect for a point crystal and point detectors an effect of about 13 percent on the basis of the assumptions of Chapter IV. This estimate does not include higher momentum effects such as core annihilation.

B. Experiment

For the two runs described below, the discriminator threshold settings of the pulse shapers were about 0.6 volts. This voltage corresponds to a gamma-ray energy of about 0.3 Mev so that each gamma-ray expending more than 0.3 Mev of energy in one of the NaI(TI) crystals gave rise to a 25 nsec output pulse from the associated shaping circuit.

The source strength at the beginning of each run was about 50 mCi of positron activity, the duration of each run being about four half-lives or about 50 hours. For 50 mCi of positron activity the random coincidence rate will be negligible (~0.1 percent of the true coincidence rate). No attempt was made to measure this rate since the half-life of the source was so short (12.9 hours). The background coincidence rate (i.e. the rate with the radioactive copper crystal absent) was measured and found to be about two counts per hour. This rate is negligible compared to the true coincidence rate, except near the end of the runs where it contributed about 0.4 percent to the true coincidence rate.

The axis defined by the center of mass of the copper crystal and the centers of the "point" detector faces was vertical so that alignment of the crystal and detectors could be achieved by means of a simple plumb bob arrangement. The error in this alignment was estimated to be less than 1 mm. This residual misalignment affects the coincidence count rate by less than 0.1 percent.

The high purity copper single crystal used in the present work was obtained from Metals Research Ltd. of Cambridge, England and was thermal neutron irradiated at the Chalk River facilities of Atomic Energy of Canada Ltd. The {III} directions of the crystal were determined by standard x-ray techniques at the Department of Metallurgy x-ray facilities before the neutron irradiations were performed.

C. <u>Results</u>

To test the collinear point geometry method two runs were made. The results are shown in Figure 21. In the first run, an exploratory one, most points were taken with only 2.5 percent statistics, making it necessary to pair points in order to reduce the error (by $\sim 1/\sqrt{2}$). However, in the second run a smaller angular range was covered, making possible better statistics. The results of the second run are also shown in Figure 21. In the results for the second run most points represent about 6400 counts thus giving 1.25 percent statistics, an improvement by a factor of two over the first run. In this second run the relative orientation of crystal and holder differed from that of the first run by about 60° . The peak in the counting rate curve was also shifted by about 60° , as expected. It is seen that the results presented are consistent with an effect of about six percent. At the left hand side of the curve for the first run can be seen some evidence of the expected 120⁰ periodicity. The curves shown in Figure 21 have been corrected for decay of the source $(T_{1/2} = 12.9 \text{ hours})$ and for background coincidences. All readings were taken at room temperature.

If one assumes a uniformly dense Fermi volume and assumes that higher momentum effects can be neglected, then the peaks in the curves shown above should be a measure of the diameter of the Fermi volume in the


79

ί.

 $\{111\}$ directions. The width of these peaks should be a measure of the diameter of the "necks" that occur in the $\{111\}$ directions. The discussion below indicates that this simple interpretation of the results is consistent with the known dimensions of the copper Fermi surface.

D. Interpretation of the Results

1. Effects of Finite Source and Detector Size

With the present experimental configuration the change in coincidence counting rate due to the presence of the "necks" of the Fermi surface will be substantially reduced from the expected 13 percent because of the finite crystal and detector size. In order to estimate the expected size of this effect, the experimental Fermi surface as given in Table I can be roughly approximated by a sphere ("the belly") of radius p₀ upon which are mounted eight "necks" which are truncated cones. The relevant dimensions of such a cone are indicated in Figure 22. The finite detector and crystal size can be approximately accounted for by use of a resolution or "density" function.



Figure 22: Fermi Surface Neck Details

By the use of this density function the "Mass" or effective volume of appropriate regions of momentum space can be found and an estimate made of the expected coincidence counting rate change.

This is done by performing an elementary volume integration over a cylinder containing a neck at each end and then comparing the result with that obtained from a similar integration performed in a direction in which there are no necks. Letting the effective volume of a cylinder in a direction in which there are no necks be $2m_1$ and that of a pair of necks be $2m_2$, it is seen that a measure of the relative count rate variation is given by the ratio m₂ where

$$m_{1} = \int_{0}^{2\pi} \int_{0}^{p_{0}} \int_{0}^{r} \int_{0}^{r} f(r) dr dz de and$$

$$m_{2} = \int_{0}^{2\pi} \int_{0}^{h} \int_{0}^{r} f(r) dz dr de + 2\pi \int_{0}^{r} f(r) [h + (b - r) + and] dr$$

$$P(r) is the resolution function$$

and P(r) is the resolution function.

2. <u>Resolution Function for Finite Detectors and Point Crystal</u>

For the case of circular detectors of diameter d and a "point" crystal source the resolution function is

$$f'(r) = 1 - \frac{Dr}{d}$$
 $r < \frac{d}{D}$

(5-2)

$$f(r) = 0 \qquad r > \frac{d}{D}$$

where D is the source to detector distance. This expression for $\rho(r)$ was obtained as a generalization of a numerical example; an analytic derivation appears to be quite difficult. For the present experimental arrangement one has $\frac{d}{D} = 1.70 \times 10^{-3}$. If one now chooses (Chapter I)

$$p_{o} = 5.10 \text{ mc} \times 10^{-3}$$

$$b = 1.00 \text{ mc} \times 10^{-3}$$

$$h = p_{m} - p_{o} = 0.70 \times 10^{-3} \text{mc}$$

$$\kappa = \frac{\pi}{4}$$
(5-3)

equations (5-1) and (5-2) yield $m_2 = 0.12$. Here p_0 is taken to be the $m_1^2 = m_1^2$ average belly radius (Chapter I). It is interesting to note that if the necks are taken to be cylinders of radius b instead of truncated cones, the effect drops to $m_2 = 0.087$, a substantial change.

3. Resolution Function for Finite Detectors and Finite Crystal

m₁

Since the copper crystal used in the present work was a cylinder whose axis was inclined at an angle of 70.5° with the line defined by the detectors, it is difficult to give a simple analytic expression for the resolution or density function. A reasonable first choice of resolution function would appear to be the following:

$$\int^{\rho} (\mathbf{r}) \, \boldsymbol{\alpha} = e^{\frac{\mathbf{r}^{2}}{2b}^{2}}$$
(5-4)

where \S is an effective source dimension (as "seen" by a detector). This function is similar to the resolution function used by Lang (1956) for correction of angular correlation results (wide slit geometry) in which the ratio of source width to detector slit width was 0.5. If, for the present arrangement one assumes an effective source size of $\$ \approx 4$ mm one finds $\mathring{r}(r) \approx e \quad \frac{-r^2}{2}$

Using this resolution function and the parameters (5-3) one obtains $\frac{m_2}{m_1} = 0.082$ which is somewhat smaller than the previous value 0.12 obtained assuming negligible source size. Including, now, for variations in b permitted by the present low-temperature data (Table I), the resultant uncertainty in $\frac{m_2}{m_1}$ is 0.004. The dependence of the calculated values of

 $\frac{m}{m_1}^2$ on other parameters such as the value chosen for \propto is illustrated by repeating the calculation for $\propto = \frac{\pi}{3}$. The calculation yields $\frac{m_2}{m_1} = 0.072$ ± 0.004 . Thus the present method should be quite sensitive to the detailed shape of the Fermi surface.

It should be noted that the experimental results are consistent with a considerable number of resolution functions. For example the resolution functions $\int (r) \propto e^{-\frac{r^2}{2}}$ and $\int (r) \propto e^{\frac{-r^2}{4}}$ give values of $\frac{m_2}{m_1}$ of 0.082 and 0.050 respectively both of which are consistent with the results shown in Figure 21. The full width at half maximum of the corresponding angular correlation curves is 32° and 45° respectively (Appendix B) however; so that the second resolution function may give a better fit to the data shown in Figure 21. Despite this difficulty, it is seen that the experimental results are consistent with a model of the Fermi surface in which "necks" occur in the $\{111\}$ directions, subtending an angle of $\lambda \approx 20^\circ$ at $\vec{k} = 0$ (see Table 1). This angle is defined by the (circular) region of contact of the Fermi surface with the Brillouin zone boundary: $\lambda = 2 \tan(\frac{r}{k_{\rm H}})$ where k_r is the neck radius at the zone boundary and $k_{\rm H}$ is the distance of this zone boundary from $\vec{k} = 0$.

Thus, taking into consideration the uncertainties in the available

Fermi surface dimensions (Table I) and in the resolution function one is led to expect an effect of about five to eight percent in the present work if the Fermi volume is assumed to be uniformly dense and if higher momentum effects can be neglected. Although this is consistent with the results shown in Figure 21, it is evident that a more accurate knowledge of the resolution function is essential if the method is to be ${}^{of}_{\lambda}$ quantitative value. It is believed that a more convenient choice of crystal orientation should permit a good estimate of the resolution function, thus obviating this source of difficulty.

E. Interpretation of the Results

The above interpretation of the results neglected complications due to higher momentum effects and ion core annihilation contributions. However, the higher momentum components associated with the contact of the Fermi surface with a hexagonal zone face will not cause difficulty since the affected momentum states will usually be merely translated by a reciprocal lattice vector that lies essentially along the axis of the sampling cylinder. For other directions normal to a zone boundary, a similar consideration applies. However, if the cylinder axis is not approximately perpendicular to a zone face small losses in the counting rate will occur. Since in copper the nonhexagonal zone faces are relatively distant from the center of the zone, it appears reasonable to assume that this contribution will be fairly small and will probably be masked by the much larger core annihilation contribution.

By use of the results on core annihilation given by Berko and Plaskett (1958) an estimate of the core annihilation contribution can be made. This estimate is made by approximating the core annihilation angular correlation curve by a parabola and considering this parabola to correspond to a

Fermi surface with $p_0 = 18$ mc x 10^{-3} . By properly weighing the contribution of the "ion core Fermi sphere" with that of the (concentric) Fermi sphere associated with the conduction electrons, it can be roughtly estimated (Appendix C) that core annihilation should lower the expected effect of 8.2 percent to about 6 percent. This is not inconsistent with the present results. A more detailed calculation does not appear to be justified by the statistics or resolution of the present work. In addition, recent calculations by Carbotte (1966) indicate that core annihilation calculations may be considerably more difficult than earlier believed.

F. Accuracy Attainable with the Method

It is perhaps of interest to consider the ultimate accuracy attainable with the present method, in order that it may be compared with the other methods used for Fermi surface determination. Various aspects of this topic are discussed below.

1. Resolution and Counting Rate

An important limiting factor in the present method is the low counting rate. In order to improve the resolution by a factor of two (i.e. to reduce the half-width of the resolution function by a factor of two) it is necessary to reduce the detector solid angle by a factor of four. This reduction in detector solid angle reduces the coincidence counting rate by a factor ~ 16 . This estimate is for a "point" crystal. For a finite crystal the decrease would be even more severe. Thus, in order to improve the resolution by a factor of two, with the rest of the experimental arrangement unchanged, the source strength used in the present work would need to be increased from ~ 50 mCi to about a Curie. Problems associated with the handling of such sources would become much more severe than those encountered so far. However, a more important limitation arises from considerations of specific activity. With presently available thermal neutron fluxes at AECL, Chalk River, it would be difficult to obtain a positron activity of much more than a Curie in the present crystal.

Other possible improvements involve an increased detection efficiency. This could be accomplished by lowering the discrimination levels of the pulse shapers and by using larger Nal(Tl) crystals. It may then be possible to improve the individual detector efficiency of 0.45 by a factor ~2 so that the coincidence count rate could be improved by a factor 4. It is thus evident from this discussion that the resolution (using the present crystal as a source) cannot be improved by more than a factor ~3 if reasonable (less than 40 feet) source to detector distances are to be used. An appropriate long-lived source, of course, would be prohibitively expensive, at present.

2. <u>Stability</u>

Another important limiting factor is the mechanical and electrical stability of the arrangement. The system could be made sufficiently stable mechanically so that the uncertainty in the coincidence counting rate could be reduced to less than 0.1 percent. However, the uncertainty in the co-incidence counting rate due to the instability of the electroncis cannot be reduced below ~ 0.1 percent without greatly increased complexity of instrumentation. Thus, for coincidence count rates in the 1 to 10 sec⁻¹ range, counting statistics would appear to set the practical limit on the precision attainable. Allowing, at most, a few hours per point, total counts of no more than a few tens of thousands could be obtained leading to statistical uncertainties ~ 1 percent per point.

G. Discussion

By decreasing the solid angle associated with a detector by a factor of four and increasing the detector efficiency by a factor ~ 2 and using the maximum available thermal neutron fluxes it should be possible to improve the resolution of the present arrangement by a factor of two while maintaining statistics of ~ 1 percent. The electronic drift could be reduced to ~ 0.1 percent by use of recycling procedures. In addition, a more convenient choice of crystal orientation should permit a good estimate of the resolution function. Thus, in the absence of core annihilation effects and other higher momentum effects, the present technique would be competitive with the other methods (Table I) except perhaps for the de Haas van Alphen effect with which, for example, the Fermi surface of potassium was found to be spherical to ~ 0.1 percent (Shoenberg, 1965).

If careful measurements, for various crystal orientations, are made with such improved resolution it may be possible to separate the coincidence counting rate contribution due to annihilation of positrons with electrons of the ion cores from the coincidence counting rate due to annihilation of positrons with valence electrons. If such a separation proves possible, it may permit measurements of Fermi surface diameters to an accuracy of ~1 percent in certain alloys composed of such tractable metals if certain assumptions can be made about the effect of alloying on core annihilation of the constituent metals. It might also be possible to investigate higher momentum effects associated with the localization of the positron in the periodic crystal potential or it may be possible to examine higher momentum effects associated with proximity of the Brillouin zone boundaries.

H. Conclusions

By use of a new positron annihilation technique employing "collinear point geometry" the Fermi surface of copper was found to be anisotropic at room temperature. The results of the present work are consistent with the picture of the Fermi surface having "necks" whose contact with the hexagonal faces of the first Brillouin zone subtends an angle of about 20° at the origin of K-space as indicated by the more accurate work done by other workers near absolute zero.

The results obtained in this work are not of sufficient precision to indicate the extent of the contribution from core annihilations. However, with better statistics and resolution, a more detailed treatment of the contribution of core annihilations will be required before further details of the Fermi surface topology can be ascertained.

Since the statistics and resolution can be improved substantially over those employed in the present work it should be possible to study, in detail, over a much larger temperature range than is the case for other methods, the Fermi surface of various metals. In addition it should be possible to apply the method to a systematic study of alloys. This is of considerable interest since little work has been done on the Fermi surface of alloys.

In addition to its promise as a tool for the investigation of Fermi surfaces, the method of collinear point geometry may prove to be of some value in the study of core electron annihilation. With improved resolution and statistics it may be possible to separate the contribution to the coincidence counting rate due to core electrons from the contribution arising from the conduction electrons. This would make possible an experimental test of core annihilation calculations.

In closing it should perhaps be noted that very little work on Fermi surfaces has been done by means of point geometry. In view of the fact that the preliminary results of Fujiwara (1965) obtained by use of noncollinear point geometry show much more structure than do those obtained by Berko and Plaskett (1958) using the wide slit method, the noncollinear point geometry method may prove to be a useful complement to the collinear point geometry method.

Finally, it can be noted that the advent of lithium drifted germanium detectors (Dearnaley and Northrop, 1966) possessing excellent energy resolution: may make possible further improvements in the point geometry method. For example, by combining some Doppler shift discrimination together with the usual angular selection, one could achieve a system with very much more sensitivity to the Fermi surface topology since the region of momentum space sampled by the proposed method could be made very small. Thus, for the collinear case, one could sample a cylinder in momentum space extending from $p_2 = \alpha p_0$ to $p_2 = \beta p_0$ instead of from $p_2 = -p_0$ to $p_2 = +p_0$. Here α and β are positive constants and p_0 is the Fermi momentum. It is seen that the method holds promise for the study of higher momentum components ($p > p_0$) since α and β could be chosen to be greater than unity.

APPENDIX A

SOLUTION OF ABEL'S INTEGRAL EQUATION

The solution of Abel's integral equation

$$S(x) = \int_{a}^{x} \underbrace{u(y) dy}_{a} \qquad 0 < x < 1, \qquad f(a) = 0$$

is given (Bocher, 1929) by

$$u(x) = \frac{\sin \pi \alpha}{\pi} \quad \frac{d}{dx} \int_{a}^{x} \frac{S(t)dt}{(x - t)'} \qquad (A-1)$$

It is desired to find the solution of the related integral equation

$$f(x) = \int_{x}^{a} \frac{g(t)dt}{(t^{2} - x^{2})^{\frac{1}{2}}}$$

Let
$$w = -x^2$$
, $v = -t^2$ and $dv = -2tdt$ and consider
 $r(-x^2) = r(w) \equiv -f(x) = \int_{a}^{x} \frac{q(t)dt}{(t^2 - x^2)^2} = \int_{a}^{w} \frac{G(v)dv}{(w - v)^2}$ where
 $G(v) = \frac{q(t)}{-2t} = \frac{q(\sqrt{-v})}{-2\sqrt{-v}}$ (A-2)

Application of (A-1) gives

<u>d</u>₩ dw

$$G(w) = -\frac{1}{\pi} \frac{d}{dw} \int_{-a^{2}}^{w} \frac{r(v)dv}{(w - v)} = -\frac{1}{2\pi x} \frac{d}{dx} \int_{-a^{2}}^{w} \frac{r(v)dv}{(w - v)^{\frac{1}{2}}} \text{ since}$$

$$= \frac{d\psi}{dx} \frac{dx}{dw} = -\frac{1}{2x} \frac{d\psi}{dx}$$

Substitution for v and w yields

ł

$$G(w) = -\frac{1}{2\pi x} \frac{d}{dx} \int_{a}^{x} \frac{\left[-f(t)\right]\left[-2t dt\right]}{\left(t^{2} - x^{2}\right)^{\frac{1}{2}}}$$
(A-3)

From (A-2) one has $G(w) = \frac{q}{-2} \sqrt{-w} = \frac{q(x)}{-2x}$

Combining this last equation with (A-3) then gives the desired result

$$g(x) = -\frac{2}{\pi} \frac{d}{dx} \int \frac{t f(t) dt}{(t^{*} - x^{*})^{\frac{1}{2}}}$$

APPENDIX B

EXPECTED ANGULAR CORRELATION CURVE WIDTH

Certain geometric relations needed for the calculation will be first outlined. Consider the rotation, through an angle ϕ , of a vector \vec{p} about the p axis. For the two positions (1 and 2) one has from the adjacent figure that

$$\vec{p}_{1} = (p_{o} \sin\psi, 0, p_{o} \cos\psi)$$

$$\vec{p}_{2} = (p_{o} \sin\psi\cos\phi, p_{o} \sin\psi\sin\phi, p_{o} \cos\psi)$$
(B-1)
(B-1)

The perpendicular distance from point \vec{p}_1 to the line defined by \vec{p}_2 can be obtained after determination of the constant α in the equation

$$(\alpha \vec{p}_{2} - \vec{p}_{1}) \cdot \vec{p}_{2} = 0$$
 (B-2)

Solving for the perpendicular distance $d_1 = |\alpha \vec{p}_2 - \vec{p}_1|$ one finds $d_1 = p_0 \sin \psi \sqrt{1 + \cos^2 \psi} - \cos \phi (\cos \phi \sin^2 \phi + 2 \cos^2 \psi)$

In order to calculate the shape (hence the halfwidth of the angular correlation curve several approximations are made:

- 1. The Fermi surface "necks" are assumed to be approximated by cylinders of height h and diameter $a_{yr} = b + \frac{h}{2}$. Here b and h have the same meaning as in Chapter V.
- 2. An effective distance (in momentum space) d_{\perp} is used. This distance is the distance between the tips of the vectors \vec{p}_{1}

and p_2 . The quantity p_0 is assumed to be given by $p_0 = p_F$ + $\frac{h}{2}$ where p_F is the magnitude of the Fermi momentum. 3. Core annihilation and other higher momentum effects are neglected.

With the above simplifications, the problem reduces to the calculation of the "mass" or effective colume of a cylinder of base a and height h if its density varies as $f(\tilde{r}) \propto e^{-gr^2}$. This density is assumed to be independent of h. The (circular) base of the cylinder rests on the $p_x - p_y$ plane, its center lying on the p_x axis a distance d_1 from the origin. The equation of the circle is

$$r = r_{\perp} = d_{\perp} \cos \Theta + \sqrt{a_{\mu}^2 - d_{\perp}^2 \sin \Theta} \quad \Theta = \sin^{-1} \left(\frac{a_{\mu}}{d} \right)$$
$$r = r_{\mu} = d_{\perp} \cos \Theta + \sqrt{a_{\mu}^2 - d_{\perp}^2 \sin \Theta} \quad \Theta = \sin^{-1} \left(\frac{a_{\mu}}{d} \right)$$

Here di > a

÷.,

The effective volume of the cylinder (i.e. a "neck") is $m(\phi) = 2\hbar \int_{0}^{\infty} \int_{r_{2}}^{r_{r}} f(r) r dr d\theta$

and the angular correlation curve is given by

$$\mathcal{E}(\phi) = \frac{m(\phi)}{m(o)} R$$

where R = $\frac{m_2}{m_1}$ is the coincidence count rate anisotropy expected at $\phi = 0$

(equation (5-1)).

A choice of $g = \frac{1}{2}$ gives a curve with height $\frac{m}{m_1} = 0.082$ and a full width at half maximum of about 32° . For $g = \frac{1}{4}$ one obtains 0.050 and 45° respectively. It would thus appear that better agreement with the ex-

perimental curves is obtained if one chooses the resolution function $r^{-r^{*}}_{3}$ (see also Chapter V) since on **e** then obtains $m_{2} = 0.062$ m_{1}^{2}

and $W_1 \approx 40^\circ$, in fair accord with the experimental curves.

APPENDIX C

EFFECT OF CORE ANNIHILATION

If it is assumed that the core electrons can be roughly described by a sort of "ion core Fermi sphere" with radius p_{ξ} and the conduction electrons by a (concentric) Fermi sphere of radius p_{ξ} one has, for wide slit geometry, for the ratio of the coincidence counting rate contributions at z = 0

$$\frac{N \text{ cond}}{N \text{ core}} = \frac{h_1}{h_2} = \frac{\int 2\pi r \, dr}{\int 2\pi \delta r \, dr} = \frac{p_F^2}{\delta P_F^2}$$

where § is the momentum space density (assumed constant) for the core electrons. Letting m₁ be the belly mass contribution, m₂ the neck mass contribution (Chapter V), and m₃ the contribution of the ion core electrons, it is seen that for point geometry the coincidence counting rate anisotropy is reduced from m₂ to m₂ $\begin{bmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 & 1 \\ m_1 & m_1 & m_1 & m_1 & m_1 \end{bmatrix} = m_2 \begin{bmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & 1 & 1 \\ m_1 & m_1 & m_1 & m_1 & m_1 \end{bmatrix}$

From Berko and Plaskett's paper one can estimate $\frac{h_2}{h_1} = 1.15$ and $p_E \sim 18 \times 10^{-3}$ mc so that $\frac{1}{1 + \frac{m}{m_1^3}} = 0.75$

Thus core annihilation would be expected to affect the coincidence count rate anisotropy by about 25 percent.

BIBLIOGRAPHY

- ANDERSON, C. D., (1932), Phys. Rev., <u>41</u>, 405.
- ANDERSON, P. W., (1963), Concepts in Solids, Benjamin, New York.
- BASSON, J. K., (1954), Phys. Rev., <u>96</u>, 691.
- BECKER, R., and SAUTER, F., (1964), <u>Electromagnetic Fields and Interactions</u>, Vol. I, Blackie, London.
- BENEDETTI, S., DE, COWAN, C. E., KONNEKER, W. R., and RPIMAKOFF, N., (1950), Phys. Rev., <u>77</u>, 205.
- BERINGER, R., and MONTGOMERY, C. G., (1942), Phys. Rev., <u>61</u>, 222.
- BERKO, S., and HEREFORD, F. L., (1956), Rev. Mod. Phys., <u>28</u>, 299.
- BERKO, S., and PLASKETT, J.'S.', (1958), Phys. Rev., <u>112</u>, 1877.
- BERKO, S., (1962), Phys. Rev., <u>128</u>, 2166.
- BLACKETT, P. M. S., and OCCHIANLINI, G. P. S., (1933), Proc. Roy. Soc. (London) <u>A139</u>, 699.
- BLOCH, F., (1928), Z. Physik, <u>52</u>, 555.
- BOCHER, M., (1929), <u>An Introduction to the Study of Integral Equations</u>, Cambridge, London.
- BOHM, H. V., and EASTERLING, V. J., (1962), Phys. Rev., <u>128</u>, 1021.
- BROWN, G., (1964), <u>Unified Theory of Nuclear Models</u>, North Holland, Amsterdam.
- BURDICK, G. A., (1961), Phys. Rev. Letters, <u>7</u>, 156.
- BURDICK, G. A., (1963), Phys. Rev., <u>129</u>, 138.
- CALLAWAY, J., (1958), Solid State Physics, 7, 99.
- CARBOTTE, J. P., and KAHANA, S., (1965), Phys. Rev., <u>139</u>, A213.
- CARBOTTE, J. P.¹, (1966), Phys. Rev., <u>144</u>, 309.
- COLOMBINO, P., FISCELLA, B., and TROSSI, L., (1963), Nuovo Cimento, <u>27</u>, 589.
- CORNWELL, J. F., (1964), Phys. Kondens. Materie 1, 161.

DANIEL, E., and VOSKO, S. H., (1960), Phys. Rev., 120, 2041.

DEARNALEY, G., and NORTHROP, D. C., (1966), <u>Semiconductor Counters for</u> <u>Nuclear Radiations</u>, E & F. N. Spon Ltd., London.

DEUTSCH, M., (1953), Progr. Nucl. Phys., <u>3</u>, 131.

DIRAC, P. A. M., (1930), Proc. Camb. Phil. Soc. 26, 361.

DONAGHY, J. J., STEWART, A. T., ROCKMORE, D. M., and KUSMISS, J. H., (1965), in: <u>1Xth Intern. Conf. Low-Temp. Phys.</u>, Plenum Press, New York.

EVANS, R. D., (1955), The Atomic Nucleus, McGraw-Hill, New York.

FOCK, V., (1930), Z. Physik, <u>61</u>, 126.

FUJIWARA, K., (1965), J. Phys. Soc. Japan, <u>20</u>, 1533.

GRAHAM, R. L., and STEWART, A. T., (1954), Can. J. Phys. <u>32</u>, 678.

- GREEN, J., and LEE, J., (1964), <u>Positronium Chemistry</u>, Academic Press, New York.
- GUSTAFSON, D. R., MACKINTOSH, A. R., and ZAFFARANO, D. J., (1963), Phys. Rev., <u>130</u>, 1455.
- HARRISON, W.¹ A.¹, and WEBB, M. B.¹, eds, (1960), <u>The Fermi Surface</u>, Wiley, New York.

HARTREE, D. R., (1928), Proc. Camb. Phil. Soc. <u>24</u>, 89.

HARTREE, D. R., (1957), <u>The Calculation of Atomic Structures</u>, Wiley, New York.

HATANO, A., KANAZAWA, H. K., and MIZUNO, Y., (1965), Progress of Theoretical Physics, <u>34</u>, 875.

HEINE, V., (1960) Group Theory in Quantum Mechanics, Pergamon, New York.

HEITLER, W., (1954) The Quantum Theory of Radiation, Oxford University Press.

HYLLERAAS, E. A., (1947), Phys. Rev., 71, 491.

HYLLERAAS, E. A., and ORE, A., (1947), Phys. Rev., <u>71</u>, 493.

- JAUCH, J. M., and ROHRLICH, F., (1955), <u>The Theory of Photons and Electrons</u>, Addison-Wesley, Reading, Mass.
- JOSEPH, A. S., and THORSEN, A.C., (1964), Phys. Rev., 134, A979.

KAHANA, S., (1960), Phys. Rev., <u>117</u>, 123.

KAHANA, S., (1963), Phys. Rev., <u>129</u>, 1622.

KANAZAWA, H., OHTSUKI, Y. H., and YANAGAWA, S., (1965), Phys. Rev., 138 A1155. KITTEL, C., (1956), Solid State Physics, Wiley, New York. KLEMPERER, 0., (1934), Proc. Camb. Phil. Soc., 30, 347. KUGEL, H. W., FUNK, E. G., and MIHELICH, J. W., (1966), Physics Letters, 20, 364. LANG, L. G., (1956), Ph. D. Dissertation, Carnegie Inst. of Tech., (unpublished). LANG, L. G., and BENEDETTI, S., DE, (1957), Phys. Rev., 108, 964. LEE-WHITING, G. E., (1955), Phys. Rev., <u>97</u>, 1557. LIVINGSTONE, W. R., (1963), Private Communication. MAJUMDAR, C. K., (1965), Phys. Rev., 140, A227. MESSIAH, A., (1962), Quantum Mechanics, Vol. II, Wiley, New York. MOHOROVICIC, S., (1934), Astron. Nachr. 253, 94. MORSE, R. W., (1960), in: The Fermi Surface, eds. W. A. Harrison and M. B. Webb, Wiley, New York. MORTON, V. M., (1960), Ph. D. Thesis, Cambridge University. NEAMTAN, S. M., DAREWYCH, G., and OCZKOWSKI, G., (1962), Phys. Rev., 126, 193. PINES, D., (1955), Solid State Physics, 1, 367. PINES, D., (1956), Rev. Mod. Phys., 28, 184. PINES, D., (1963), Elementary Excitations in Solids, Benjamin, New York. PRICE, B. T., HORTON, C. C., and SPINNEY, K. T., (1957), Radiation Shielding, Pergamon, New York. PIPPARD, A. B., (1957), Phil. Trans. Roy. Soc. (London), A250, 325. RAIMES, S., (1957), Reports on Progress in Physics, 20, 1. RAIMES, S., (1961), The Wave Mechanics of Electrons in Metals, North-Holland, Amsterdam. REITZ, J. R., (1955), Solid State Physics, 1, 1. ROAF, D. J., (1962), Phil. Trans. Roy. Soc. (London), A255, 135.

- ROCKMORE, D. M., and STEWART, A. T., in: Wayne State University Positron Annihilation Conference, 1965, (To be Published).
- RUARK, A. E., (1945), Phys. Rev., <u>68</u>, 278.
- SEGALL, B., (1962), Phys. Rev., <u>125</u>, 109.
- SHOENBERG, D., (1962), Phil. Trans. Roy. Soc. (London), A255, 85.
- SHOENBERG, D., (1965), in: <u>IXth Intern. Conf. Low-Temp. Phys</u>., Plenum Press, York.
- SIMONS, L., (1953), Phys. Rev. <u>90</u>, 165.
- SLATER, J. C., (1963), <u>Quantum Theory of Molecules and Solids</u>, McGraw-Hill, New York.
- SOMMERFELD, A., (1928), Z. Phys. <u>47</u>, 1.
- STEWART, A. T., (1957), Can. J. Phys., <u>35</u>, 168.
- STEWART, A. T., (1961), Phys. Rev., <u>123</u>, 1587.
- STEWART, A. T., SHAND, J. B., DONAGHY, J. J., and KUSMISS, J. H., (1962), Phys. Rev., <u>128</u>, 118.
- STUMP, R., (1955), Phys. Rev., 100, 1256(A).
- TERRELL, J. H., BERKO, S., and WEISBERG, H. L., in: Wayne State University Positron Annihilation Conference, 1965, (To be Published).
- TINKHAM, M., (1964), Group Theory, McGraw-Hill, New York.
- WALLACE, P. R., (1960), Solid State Physics, 10, 1.
- WHEELER, J. A., (1946), Ann. N. Y. Acad. Sci., 48, 219.
- WIGNER, E., and SEITZ, F., (1933), Phys. Rev., <u>43</u>, 804.
- WILLIAMS, R. W., LOUCKS, T. L., and MACKINTOSH, A.R., (1966), Phys. Rev. Letters, <u>16</u>, 168.
- ZIMAN, J. M., (1960), <u>Electrons and Phonons</u>, Oxford University Press.
- ZIMAN, J. M., (1964), <u>Principles of the Theory of Solids</u>, Cambridge University Press.